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MERCURY EMISSIONS FROM COAL FIRED POWER PLANTS LOCAL IMPACTS ON HUMAN HEALTH RISK

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Table of Contents

List of Figures

List of Tables

1. Introduction

1.1 Background

Mercury is a neurotoxin that accumulates in the food chain and is therefore a health concern. Concentrations of mercury in the air are of little direct health concern. However, mercury in the air falls onto the Earth's surface through dry and wet deposition processes. This mercury can enter water bodies where a small percentage (< 10%) is transformed to methyl mercury. This chemical form of mercury readily enters the food chain and bioaccumulates. Upper trophic level fish can have mercury concentrations several orders of magnitude greater than that found in the water or sediment. As mercury accumulates in these organisms, ecological risks occur and potentially human health risks may occur through fish consumption.

On December 15, 2003, EPA signed its first ever proposal to substantially cut mercury emissions from coal-fired power plants. The Utility Mercury Reductions proposal would cut mercury emissions by nearly 70 percent when fully implemented. The Utility Mercury Reductions rule would permanently cap emissions from coal-fired power plants and provide companies with flexibility to achieve early reductions of mercury. EPA proposed two alternatives for controlling mercury.

- Require power plants to install controls known as "maximum achievable control technology (MACT) under section 112 of the Clean Air Act. If implemented, this proposal would reduce nationwide mercury by 14 tons or about 30 percent by early 2008.
- Or, EPA proposed a market-based "cap and trade" program that, if implemented, would reduce nationwide utility emissions of mercury in two phases. When fully implemented mercury emissions would be reduced by 33 tons (nearly 70 percent).

On February 24, 2004, EPA signed a proposed cap-and trade rule that supplements its December 15, 2003 proposal to cost effectively reduce mercury emissions from power plants. States may choose to adopt the cap-and-trade program to achieve and maintain the necessary emission budgets. Public comments were accepted until June 29, 2004. In all over 1 million comments were received. EPA is currently reviewing and addressing the comments in order to finalize the rule by March 15, 2005.

Although a Cap and Trade approach is being considered, there are technical doubts pertaining to local deposition of mercury leading to “hot spots” that would make a Cap and Trade approach unacceptable to many. This has received widespread attention in the literature (ES&T, 2004). The following are selected quotes from public health officials in the past year.

- “Unlike most pollutants, mercury is highly toxic and does not disperse easily, creating “hot spots” of contamination.” (Kathleen McGinty, Director of Pennsylvania Department of Environmental Protection, July 2, 2004 (http://www.dep.state.pa.us/newsletter/default.asp?NewsletterArticleID=8850&SubjectID=)
- “Specifically, we are concerned that local "hot spots" of elevated mercury may result or worsen, especially if the required reduction levels are not sufficiently strict.” (Renee Cipriano. Director of Illinois Environmental Protection Agency, February 26, 2004

(Testimony to the U.S. EPA regarding the U.S. Environmental Protection Agency's Proposal to Control Emissions of Hazardous Air Pollutants from Utilities (January 30, 2004, 69 Federal Register 4652) Docket ID No. OAR 2002-0056.

- "We generally support market-based approaches such as cap and trade schemes, yet we have an equally strong objection to the exclusive use of cap and trade schemes where local emissions "hot spots" are a concern. While mercury pollution and emissions are widespread, indeed a global problem, we share the concerns of many states that EPA's proposed rule understates the needs for local controls as well", letter from Stephen Mahfood, Director Missouri Department of Natural Resources to Michael Leavitt, Director U.S. EPA.
- "Sulfur dioxide is light, and travels long distances; power plants in the Midwest can cause acid rain in Maine. So a cap on total national emissions makes sense. Mercury is heavy; much of it precipitates to the ground near the source. As a result, coal-fired power plants in states like Pennsylvania and Michigan create "hot spots" – chemical Chernobyls – where the risks of mercury poisoning are severe. … That probably means thousands of children will be born with preventable neurological problems." Paul Krugman, New York Times, p. A-23, April 6, 2004.

1.2 Defining a "Hot Spot."

Although the term "hot spot" appears frequently in the health and environmental literature, precise definitions do not. A "hot spot" is a spatial anomaly, i.e., a location whose properties exceed those generally expected in the area. In statistical terms, a hotspot is an outlier whose properties exceed more than about 2 or 3 standard deviations above the relevant mean. Some authors have simply defined "hotspots" as the highest observed values; for example, Worm et al. (2003) found a range of diversities in ocean predator species of less than one order of magnitude, among which the highest groups were termed "hotspots". Lebret et al. (2000) used the term to refer to locations where the ambient air quality standard for NO_2 is routinely exceeded; in the two locations studied, the ranges of ambient values were less than a factor of 3. These two examples of "hot spots" would not meet the statistical definition.

However, the expected ranges of environmental concentrations depend heavily on the "natural" or background values and on the length of the measurement period (i.e., the averaging time). In many cases, environmental concentrations are log-normally distributed (skewed towards high values), so that the distribution is best described by exp(mean logarithm) and exp(standard deviations of the logarithms). These statistics are referred as the log-mean and the geometric standard deviation (GSD). As an example, Lu et al. (2005) studied the distribution of the polycyclic aromatic hydrocarbon naphthalene in Southern California and found a large range of values, in part because there is little or no natural background. The log-mean and GSD for 13 one- to two-month averages were 227 ng/m^3 and 1.57, leading to an expected range (GSD^4) of a factor of 6. However, for 16 four-day averages in other locations, these values were 269 ng/m^3 and 3.12, leading to an expected range of a factor of 94. In both of these distributions, the top 3 or 4 values appear to be outliers and thus bona fide "hot spots", since deleting them reduces the range of the distribution by about a factor of 2. This would not have been the case in the presence of a substantial natural background.

While small-scale transient hot spots may be a valid concern for inhalation exposures, the situation with mercury is quite different. The exposure pathway is through diet, and the relevant human exposure times relate to the development of the fetus and are of the order of months. Although a large point source of Hg indeed constitutes an emission "hot spot", it does not constitute a fetal exposure hot spot. In addition to the substantial global background in Hg air concentrations and deposition, the following processes act to smooth out spatial anomalies:

Atmospheric variability, including winds and precipitation.
Re-emission of mercury from vegetation.
Terrestrial leaching and washout in transferring watershed deposits into water bodies.
Aquatic mixing within water bodies.
Spatial and temporal variability in biomagnification processes.
Variability among fish species.

Only atmospheric variability is included in the models that may be used to define deposition "hotspots." The other processes involve spatial variability, especially with regard to mixing within the receiving waters, for which the size of the water body may be key. In order for a local Hg deposit to pose a risk to a developing fetus, its mother must routinely consume high-Hg fish from an affected water body for several months, probably at the rate of 2 or 3 meals per day. While this scenario is unlikely in any event, it also requires a substantial body of water, say of the order of tens of square km.

1.3 Statistical Methods.

Many empirical findings concerning Hg are subject to experimental error, which may be considerable in some cases. Accordingly, statistical methods may be required to gain an understanding of the data. They include averaging, correlation, and linear regression, using established relationships to try to reduce experimental variability. In all cases, "statistical significance" implies a 95% probability that the finding is not due to chance alone, denoted as "$p < 0.05$".

1.4 Contents of the Report.

This report examines the possibility that coal-fired power plants act as local sources leading to mercury "hot spots", using a three-tiered approach. First, the worldwide literature was searched for reports of deposition around mercury sources, including coal-fired power plants. Second, soil samples from around two mid-sized U.S. coal-fired power plants were collected and analyzed for evidence of "hot spots" and for correlation with model predictions of deposition. Third, a risk assessment construct was developed that demonstrates a possible approach for examining human-health risks that might be associated with local deposition of mercury emitted from coal-fired power plants. Based on this work, conclusions about the impacts of "hot–spots" are made.

The health impacts of mercury arising from coal-fired power plants comprise a complicated issue with many active areas of research. To provide context for the current studies and to keep current with the latest findings, the literature on mercury deposition, transformation and bioaccumulation in the food chain, on fish consumption, and on health effects has been followed closely. Appendix B presents a review of some of the latest findings from the literature on fish consumption, levels of mercury in fish, and modeled deposition under various regulatory control

scenarios. Appendix C presents an annotated literature review of findings on the studies of health impacts of mercury, exposures to mercury through fish consumption, atmospheric modeling of mercury transport and deposition, measured mercury data, and reviews, editorials, and opinions that have been published recently.

2. Evidence from the Literature for "Hot Spots" Near Mercury Emissions Sources

The rationale for regulating air emissions of mercury from U.S. coal-fired power plants largely depends on mathematical dispersion modeling, including the atmospheric chemistry processes that affect the partitioning of Hg emissions into elemental (Hg_0) and the reactive (RGM) forms that may deposit more rapidly near sources. Mercury is a global pollutant and therefore, modeling estimates are often based on a large scale. Fine scale, (< 20 km) modeling of point sources is not performed often. In addition, there is evidence (Edgerton 2004, EPRI 2004) that reactive gaseous mercury in coal-fired power plant plumes quickly reduces to elemental mercury. This chemical process, which would greatly affect the amount of local deposition however, the corresponding chemical reactions are not included in the available local mercury deposition models. As a result, field data are necessary to check for the existence of mercury "hot spots" near coal-fired power plants. This literature review considers the empirical support for this hypothesis. Deposition around other major sources of mercury emissions, calciners and chlor-alkali plants, is also reviewed. The extant experimental data are considered at three spatial scales: local (< 30 km), regional (< ~300 km), and national (multi-state data).

2.1 Local Evidence

The literature search found several studies dating back 30 years in which various manifestations of local Hg deposition (within a few km) were related to coal-fired power plants or chlor-alkali plants. These studies measured concentrations of Hg in soil, lake sediments, precipitation and fish. To place these results in a common framework, the relationships between background concentrations and those obtained near the plant were estimated, as well as the fractions of emitted Hg that had been deposited and retained during the period of facility operation. Where possible, these relationships were considered as a function of the receptor's distance from the plant.

2.1.1 Soil and Vegetation Studies

Mercury levels in soil and vegetation may provide a time-integrated average of deposition patterns near a mercury source. Soil and vegetation mercury concentrations will reflect background levels plus the incremental effects of local wet and dry deposition. Experimental studies (Wang et al., 2003) indicated that soils exposed to varying levels of Hg in air showed increased soil Hg concentrations with increasing air Hg concentrations. Factors complicating this paradigm include the possibility of Hg leaching through the soil and being re-emitted to the atmosphere. Gustin et al. (2004) found that photo-reduction of ionic mercury (Hg^{+2}) at the soil surface led to re-emission of mercury as Hg(0). As Hg^{+2} is a major contributor to wet deposition, this may be an important mechanism for reducing Hg soil burdens in areas of enhanced deposition.

2.1.1.1 Coal-fired Power plants

The earliest attempts at assessing Hg impacts from coal-fired power plants were based on the Hg content of surficial soil samples (Klein and Russell, 1973; Anderson et al., 1977; Crockett and Kinnison, 1979). This technique is highly dependent upon definition of the background soil content, which is not always reliable. The reported incremental Hg concentrations ranged from 29% (Klein and Russell, 1973) to 42% (Anderson et al, 1977) above background, corresponding to retention of 3-5% of cumulative emissions.

The Four Corners study (Crockett and Kinnison, 1979) measured soil concentrations at downwind distances of 1, 3, 7, 15, and 30 km at a total of 70 locations and concluded that significant accumulation of mercury was not occurring near the plant Four Corners is a large coal-fired plant (2150 MW) with two relatively short stacks (76 and 91 m). Annual rainfall is low (15 – 20 cm/yr), which limits opportunities for wet deposition. The authors did not measure background Hg but concluded that "mercury was not accumulating in the soil", based mainly on comparisons of local soil Hg concentrations with those reported in the literature for other locations. However, reanalysis of the published data on individual soil samples indicates a significant overall (log-log) slope of -0.11 ($p < 0.005$) as a function of downwind distance. Figure 1 shows the medians and ranges of soil mercury concentrations as a function of distance from the plant. The figure shows a wide variation in soil Hg at each distance, characterized by a more rapid decrease in soil Hg near the plant and the suggestion of a secondary peak at about 15 km downwind. A possible rationale might thus be a close-in peak due to RGM washout during rain events and a more distant peak due to plume touchdown and dry deposition. This secondary peak would be consistent with typical distances expected for dry deposition based on modeling. However, it is not statistically significant in this case. Various values of background Hg were assumed in order to estimate the fraction of emissions deposited and retained in the soil; the wide range of these estimates shows the sensitivity to this parameter. It appears that around 2 -12% of the plant's Hg emissions may have been deposited and retained in the soil, which would correspond to excess deposition rates of about 20 – 120 % above assumed background deposition rates. Estimates of the fractions of deposition are sensitive to the maximum downwind distance considered, which was 30 km in these analyses.

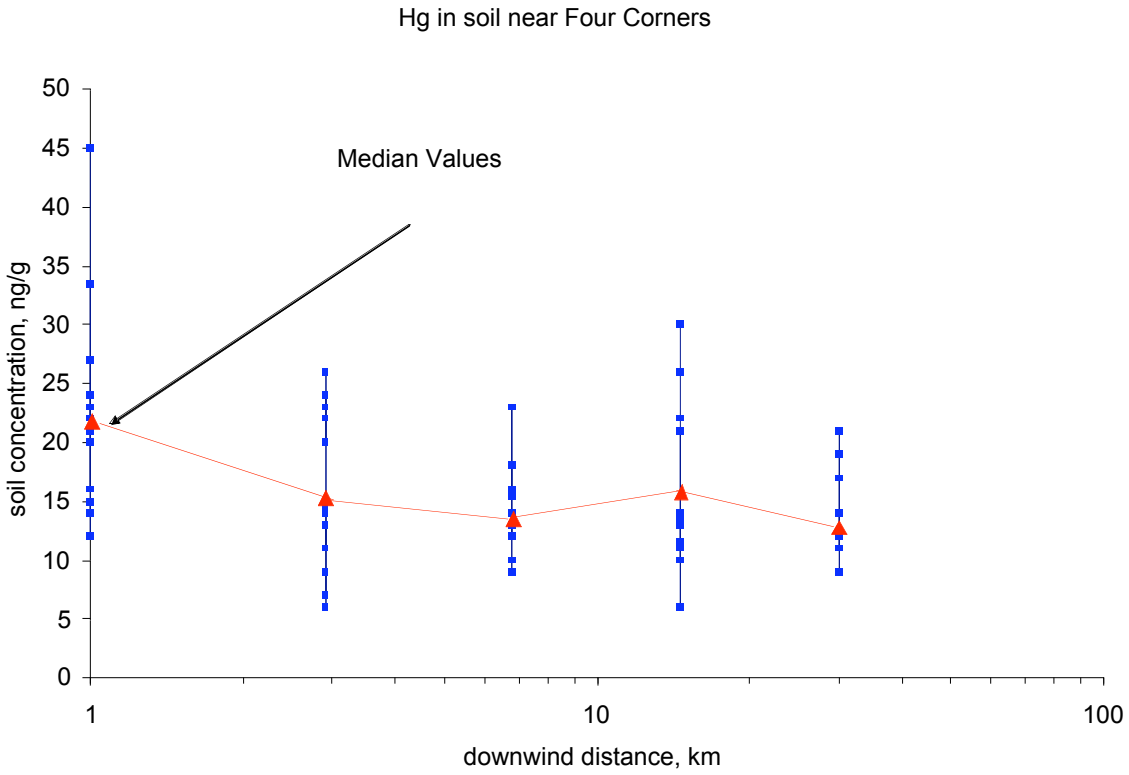

Figure 1 Soil Concentrations as a function of distance from the Four Corners power plant. (Crockett and Kinnison, 1979).

The Kincaid study (Anderson, 1977) collected data in 1973 and 1974 on airborne particulate matter, soils, sediments, and wildlife (fish and ducks) in a nearby lake. Kincaid is an 1150 MW facility that began operating in 1967. The Kincaid plant originally burned locally mined coal but switched to Powder River Basin coal in the 1990's. Soil samples were collected in April of 1974 on a 3.2 km (2 mile) grid covering 38.6 km with the power plant at the center of the grid. Two soil types were found in the sampling. North of the plant the soil was Sable silty clay loam and south of the plant the soil was a Virden silty clay loam. Ninety samples were collected with a minimum value of 1 ppb and a maximum value of 37 ppb. The mean to the south of the plant was 15 ppb while the mean to the northwest of the plant was 19 ppb and the mean to the northeast of the plant was 22 ppb. The difference between the northeast quadrant and the southern quadrants was statistically significant ($p < 0.05$) however; it may be attributable to the difference in soil types. During the 1970's, estimated mercury releases from the plant were around 430 kg/yr. Using this as a basis, it was estimated that 785 kg (18%) of the total emissions were deposited within 20 km of the plant.

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2.1.1.2 Other Major Hg Emission Sources.

Several investigators have considered mercury deposition near other types of known sources such as calciners (Abbott et al., 2003), chlor-alkali plants (Biester et al., 2002a,b; Sensen and Richardson, 2002; Lodenius, 1998), and thermometer factories (Wang et al., 2003). The study conducted by Abbott et al. examined soil samples within 5 km of a calciner that had been operating as an ore roaster for 37 years. Estimated mercury emission rates from the 75 m stack were 10 g/h with 88% as reactive gaseous mercury (RGM) and the remaining 12% as particulate mercury. Soil samples were collected on a gridded ring of 1, 2, 3, and 5 km from the source, Figure 2. Average concentrations in these rings decreased with distance from the source and were 17.6 ppb, 17.5 ppb, 15.5 ppb, and 12.3 ppb. The overall range was 7 – 23 ppb. Based on an assumed background concentration of 7 ppb, it was estimated that 2.5% of the total mercury emissions remained in the soil (Abbott, 2003). This fraction may have been larger if a larger area around the plant had been considered.

Soil studies around chlor-alkali plants show a region of excess Hg deposition extending approximately 1 – 3 km. Using lichens as a biomarker, Sensen and Richardson (2002) found decreasing levels of mercury with distance from the plant for 3.4 km. After this distance, mercury levels were consistent with background levels. Peak Hg levels were found 125 – 250 m from the plant. Soil mercury levels were elevated at distances up to 1 km downwind of three

chlor-alkali plants (Biester et al., 2002a,b). In each case, peak levels were within a few hundred meters of the plant. Calculated net deposition rates to soil within 1 km of these three plants ranged between 2300 and 8900 _g/m^2/y. Moss bags were used to estimate, a deposition rate of 480 _g/m^2/y at a distance of 200 m from another chlor-alkali plant (Lodenius, 1998). Both studies suggest local deposition rates that are substantially greater than typical background deposition rates (~10 _g/m^2/y) for wet deposition in the U.S. from the Mercury Deposition Network (MDN). Dry deposition is not accurately known, but studies near a chlor-alkali plant suggest that at least 70% of total Hg deposition is dry deposition (Lodenius, 1998). However, if this were true for ambient deposition in background areas away from sources, it would suggest 20 _g/m^2/y as a background dry deposition rate, which would have major implications on national Hg budget estimates. Thus, the estimated deposition rates within 1 km of chlor-alkali plants are as much as several hundred times background, thus comprising true hot spots. Chlor-alkali plants are known to have fugitive emission rates (non-stack releases) that are similar to stack emission rates (Southworth, 2004). It is not clear if the local effects are due primarily to fugitive or to stack releases of Hg. Near chlor-alkali plants, soil Hg concentrations are often above 1 ppm (Biester et al., 2002a,b; Southworth, 2004; Wang et al., 2003) in contrast to estimates of background concentrations of 50 – 100 ppb at these sites.

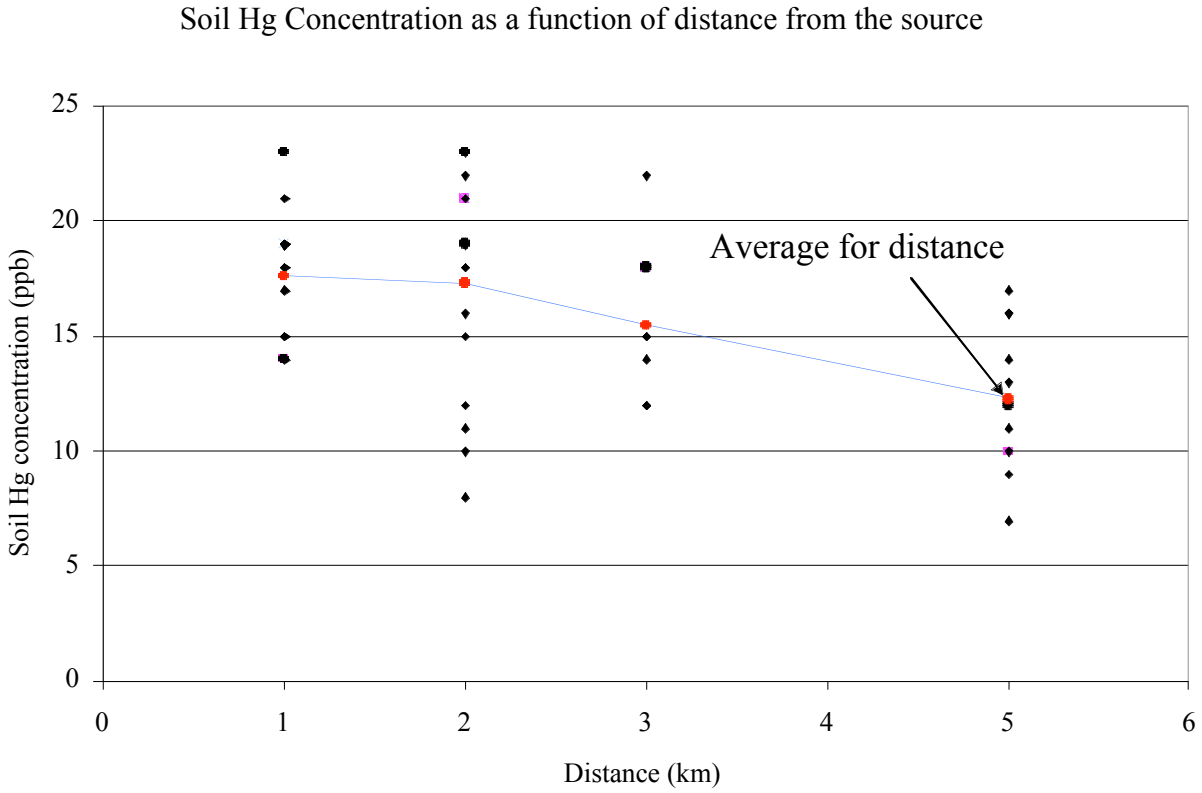

Figure 2 Soil concentration versus distance from the Calciner (source of Hg) (Abbot,2003)

2.1.2 Sediment Studies.

Three studies (Anderson et al., 1977; Kotnik et al., 2000; Menounou and Presley, 2003) include data on the Hg content of sediment cores obtained from water bodies near to coal-fired power plants. This technique has the advantage of potential relevance to impacts on fish but has the

disadvantages of uncertain time periods and the difficulty of considering decay rates in terms of downwind distance. At the Kincaid plant (Anderson et al, 1977), the excess Hg in sediments was estimated through sediment cores, and the data supported approximately 30% increase in Hg concentrations in the eight years after the plant began operations. The total mercury mass deposited was less than 1% of cumulative emissions. In Texas (Menounou and Presley, 2003), there was about 50-90% more Hg in near surface sediments from a lake a few km from the 460 MW plant than from two lakes about 30 km away. The decreases in Hg with depth within the core were also substantially larger in the sediment cores from the nearby lake. The studies at the Sostanj power plant in Slovenia (Kotnik et al., 2000) did not find evidence of increases in sediment Hg; values were comparable to other sediments (50 – 160 ppb), but background values were not obtained for comparison.

2.1.3 Precipitation Studies.

2.1.3.1 Coal-fired power plants

Two studies (Kotnik et al., 2000; Risch, 2003) considered variations in the Hg content of precipitation as a marker for local impacts of coal fired power plants. In Slovenia (Kotnik et al., 2003), it was not clear that valid annual averages had been obtained and no comparisons were made with local background. Mercury values in rain ranged from 6 – 11 ng/l, which is comparable to typical levels in the United States from the MDN. In rain, most mercury (60%) was bound to particulate matter with up to 80% associated with particulates at the stations nearest the plant. Mercury concentrations in snow were 23–54 ng/l with over 95% of the mercury associated with particulate matter.

In Indiana (Risch, 2003), the author reported no significant difference between the precipitation Hg data collected at a mercury deposition network site located 2 km from the 1300 MW Clifty Creek coal-fired power plant, as compared to 3 other mercury deposition network sites in the state. However, precipitation chemistry is frequently sensitive to the rate and thus amount of precipitation, and when this factor is taken into account in a multiple regression analysis, the site near the plant appears to have an excess Hg level of about 12% (Figure 3). With normalized deposition (deposition/precipitation) as the dependent variable, the increment is just significant ($p < 0.03$).

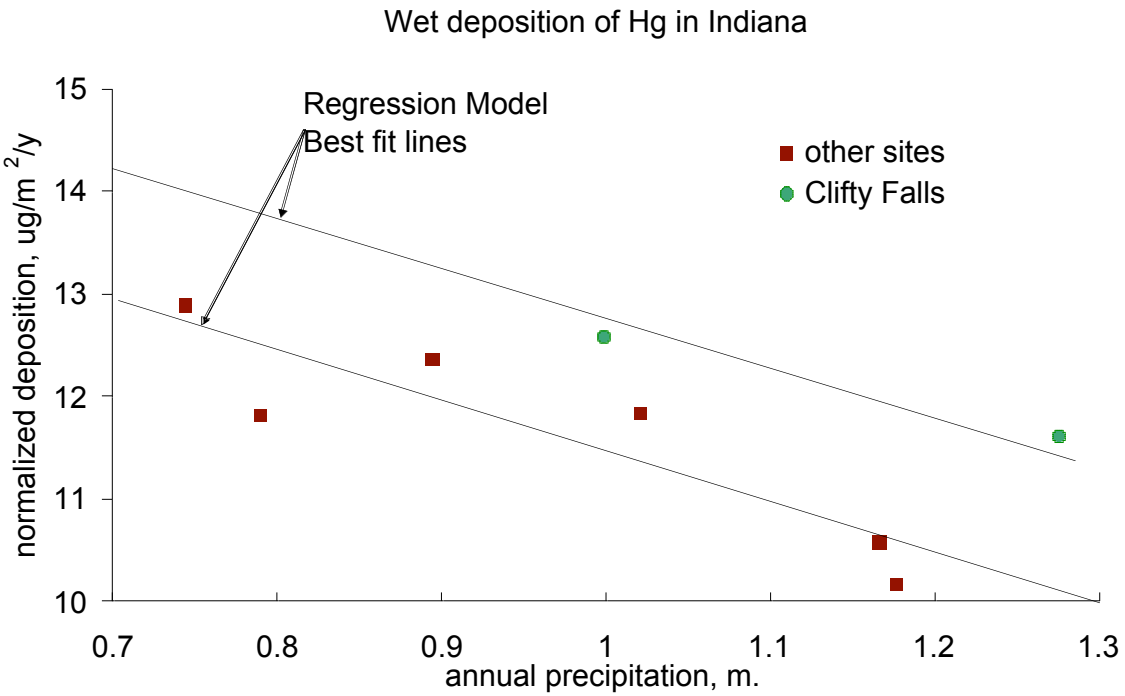

Figure 3 Mercury deposition at four Indiana MDN sites. Clifty Falls is approximately 2 km form the Clifty Creek Coal power plant.

2.1.3.2 Other Hg Emission Sources.

Mercury accumulation in snow near an ore calcining plant in southeast Idaho was studied by Susong et al. (2003). Sixty-four samples were collected with 5 km of the plant. Hg concentrations in snow ranged from 4.7 – 27.3 ng/l with concentrations generally higher near the plant. Background samples at 95 locations in the region showed total mercury concentrations ranging from 0.9 – 16.6 ng/L. This is statistically much lower than for the data near the plant. However, the interesting point of this study is that the plant was not in operation during the snow accumulation period that winter. The authors concluded that the higher values in snow near the calciner were due to re-emission from the soil, which was known to have elevated mercury levels (Abbott et al., 2003).

2.1.4 Studies of Fish Hg Content.

Two studies reported the Hg content found in various species of fish collected in or near the impacts zones of power plants. At the Kincaid plant in Illinois (Anderson et al., 1977), the fish caught near the plant had substantially less Hg than those from locations that are more distant. However, it was not clear whether this comparison included adjustments for fish size. In a study of 23 farm ponds near the Dickerson plant in Maryland (Pinkney et al., 1997), fish lengths and detailed water chemistry data were also reported, which turned out to be important. To examine these factors, the rate of wet deposition of Hg was estimated for each pond based on the modeled isopleth plots presented in the paper. In multiple regression analysis, the log of fish Hg content was significantly ($p < 0.01$) associated with the log of wet Hg deposition (coefficient = 0.5, suggesting a square root rather than a linear relationship), pond water conductivity (coefficient = -0.5), and fish length for sunfish and bass pooled (n=37). Fish length was essentially a surrogate for fish species in this study. Water conductivity, hardness, and alkalinity were highly inter-

correlated; pH was not a significant predictor of fish Hg. Water quality was not associated with estimated Hg wet deposition. Figure 4 is scatter plot of these data, based on fish Hg adjusted to a common level of pond water conductivity. Note that only three samples clearly exceed the EPA guideline for fish Hg content (0.3 ppm), notwithstanding the apparent local effects of the power plant.

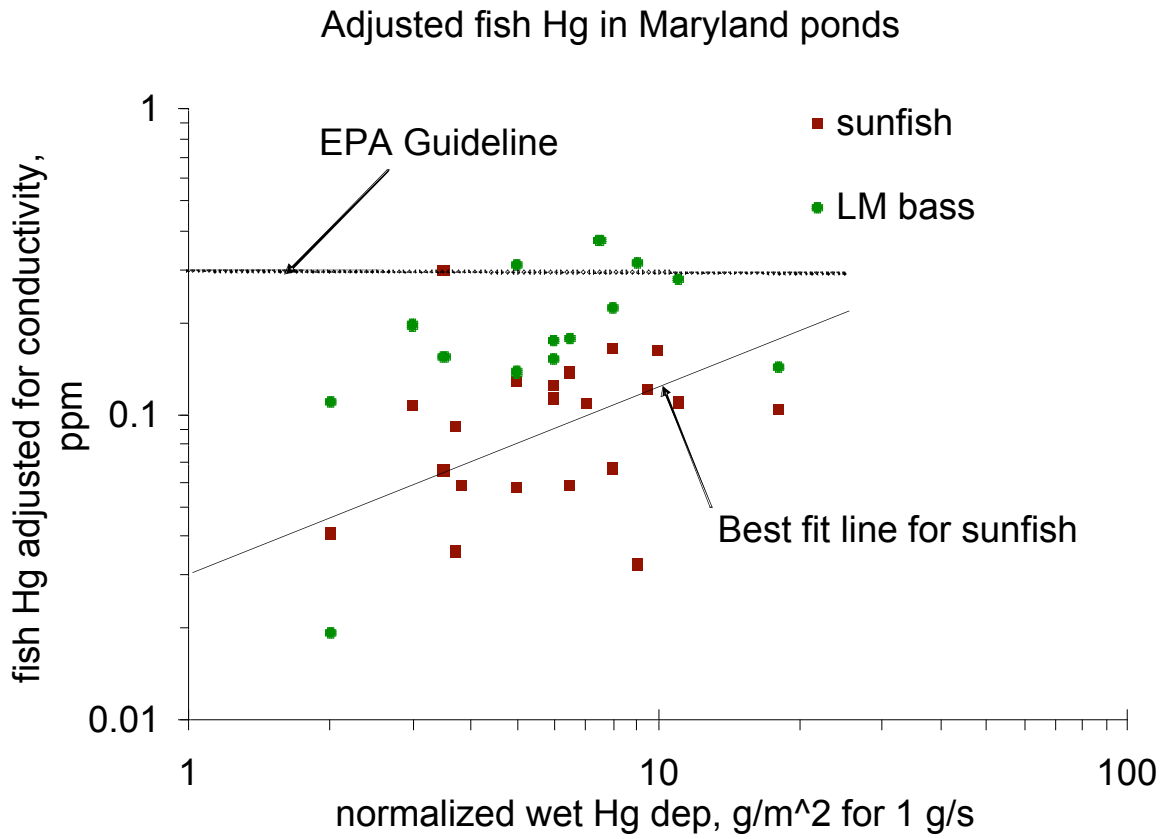

Figure 4 Correlation of fish Hg content with increased wet deposition of Hg near the Dickerson Power Plant. (data from Pinckney et al., 1997)

Table 1 summarizes the available data for near-field deposition around seven coal-fired power plants. The table contains soil, sediment, and precipitation data along with estimated percentage of mercury emissions that were deposited locally.

Table 1 Environmental data on the impacts of local Hg deposition near coal-fired power plants.

Reference	Plant data	Emissions (kg/y)	Environmental data, ppb in soils and sediments, ug/m^2/yr in precipitation, ppm in fish.				Estimated percent deposited	Remarks
			type	# samples	mean	Backgrnd *		
Klein & Russell (1973)	Campbell, MI 650 MW 122 m stack	~290	soil	90	10.2	7.9	2.7	Irregular impact area
Anderson & Smith (1977)	Kincaid (IL) 1200 MW 2 152 m stacks	531	soil	90	22	15.5	4.6	Change in soil type over sampling domain. Range 5 – 37 ppb.
			sed	36	49	37		
			fish	120	0.07	0.35		
Crockett & Kinnison (1979)	Four Corners (NM) 2150 MW 2 76, 2 91 m stacks	595	soil	70	14.5	**0	20.6	No background measurements.
						**3	16.3	
						**6	12.0	
						**9	7.8	
Pinkney et al. (1997)	Dickerson (MD) 543 MW	~240	fish	69 sunfish	0.08			See Figure 3 for data on fish Hg versus modeled deposition.
				42 LM bass	0.19			Range 0.03– 0.43 ppm.
Kotnik et al. (2000)	Sostanj, Slovenia 775 MW 100, 150, 230 m stacks	314	Pre-cip		7.4-13.7		5	No background measurements based on 20 km radius and dry dep = wet dep.
			Sed.		53-166			Higher values in surface layers.
Menounou & Presley (2003)	Gibbons Cr (TX) 460 MW	~430	Sed.	13	94	80	3	Based on 25 km radius and median excess deposition of 9.4 ug/m^2/y
Risch (2003)	Clifty Cr (IN) 1300 MW	184	Pre-cip	4 sites, 2 yrs	12.3	11		Other plants in Ohio and Kentucky not considered

* Background values in all studies are estimates and may not reflect true background. Often the lowest measured value is cited as background.

** Assumed background values for illustrative purposes. No attempt made to measure background.

2.2 Regional Data

2.2.1 Pennsylvania Wet Deposition

As an example of regional relationships, data on Hg emissions from Pennsylvania power plants (http://www.epa.gov/ttn/atw/combust/utiltox/utoxpg-old.html#DA3) were used with 1998-2002 wet Hg deposition obtained from the Mercury Deposition Network (MDN) in Pennsylvania. The 8 Pennsylvania MDN stations were operated for various portions of this period; each annual wet deposition estimate was thus treated as a separate observation (n=22). The straight-line distance between each MDN site and each of the 36 plants in Pennsylvania, whose Hg emissions ranged from 637 kg to 45 g, totaling 3622 kg, was computed from latitude-longitude data, and each emission rate was weighted by the inverse square of this distance and then summed. For the 8 MDN sites, these sums ranged from 0.09 kg/km^2 to 1.7 kg/km^2. The nearest MDN site to the largest coal-fired power plant was approximately 40 km and this MDN site tended to have the highest wet deposition.

Multiple regression analysis were then used to deduce source-receptor relationships, considering the year, site elevation, and site latitude as possible confounders. No account was taken of other point sources of Hg (such as urban areas, incinerators, or out-of-state sources) or of prevailing wind directions. MDN site latitude was included as a possible confounder because the national map of Hg wet deposition seems to show consistent increases from north to south in the eastern states. The regression coefficients for site elevation and year were far from significant and were dropped from further consideration. Latitude and (emissions/distance^2) were moderately (negatively) correlated, which posed a co-linearity problem. However the best fit to these 22 observations was obtained in a log-log regression based on the emission parameter alone, which had a log-log coefficient of 0.11 ($p<0.010$), Figure 4. This result implies that 11% of the wet Hg deposition in Pennsylvania is associated with coal-fired power plants in the state, under the modeling assumptions stated above. This relationship implies that a reduction in coal-fired power plant emissions of 50%would result in a 5.5% reduction in wet deposition of Hg.

The regression also implies that wet deposition of Hg may decrease more slowly with downwind deposition than originally assumed. Theory would suggest a decrease proportional to the inverse of the distance squared. In contrast, this empirical model would predict the following annual deposition (in $\mu g/m^2$)-distance relationship for an isolated point source of 636 kg Hg per year: 1 km., 19.5; 3 km, 15.3; 10 km, 11.7, 30 km, 9.2; 100 km, 7.1. Taking the last figure as "background", the annual wet deposition would be about 1.1% of emissions and the total Hg deposition would be about 3.3%, assuming that dry deposition accounts for two-thirds of all deposition.

2.2.2 Spanish Cattle Data

Additional information on Hg deposition at the regional scale is provided by the results of Lopez et al. (2003), who analyzed the Hg content of calf kidneys in relation to distances between the farms involved and major point sources of Hg in Northwest Spain. (e.g. a total 1200 kg/y from coal-fired power plants and similar amounts from other industries). Downwind distances ranged up to 140-200 km; the duration of this deposition was not mentioned. Based on approximations

from the scatter-plots in the paper of Lopez et al., (see Figure 6 below), it appears that deposition decreased with downwind distance to the 0.4 power for the power plants and 0.7 power for the industrial area. Neither of these slopes is significantly different from the expected square-root relationship.

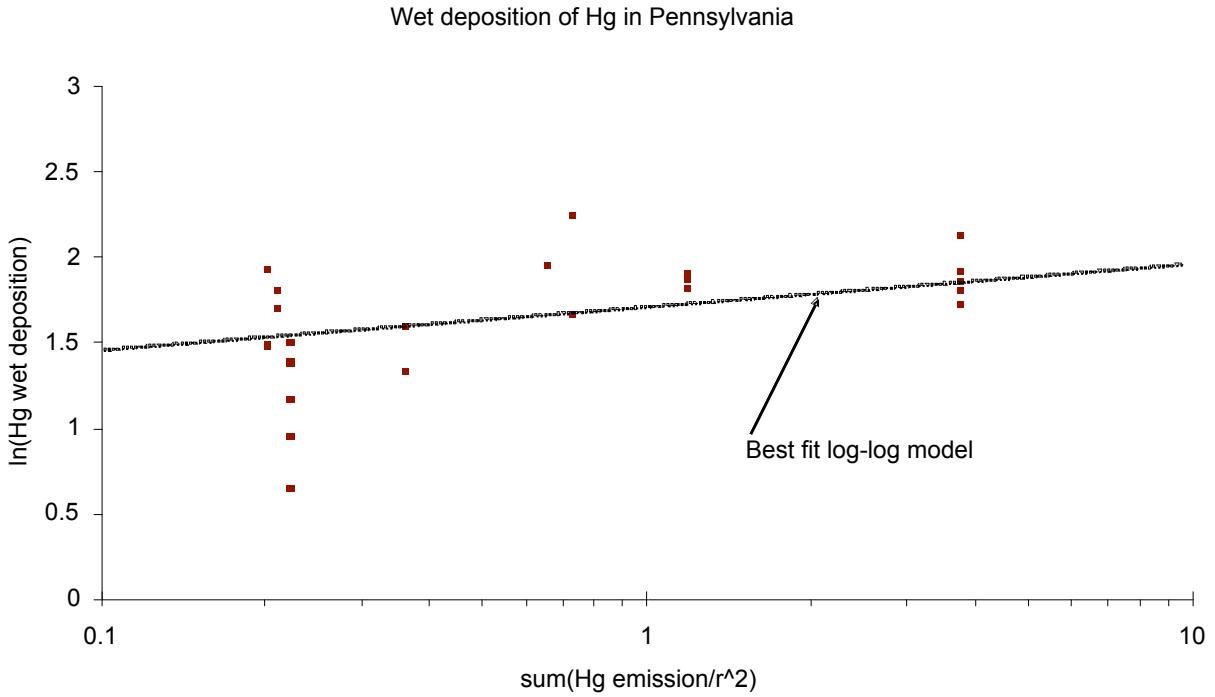

Figure 5 Log-log correlation of mercury emissions from coal-fired power plants in Pennsylvania and measured wet deposition at MDN sites in Pennsylvania.

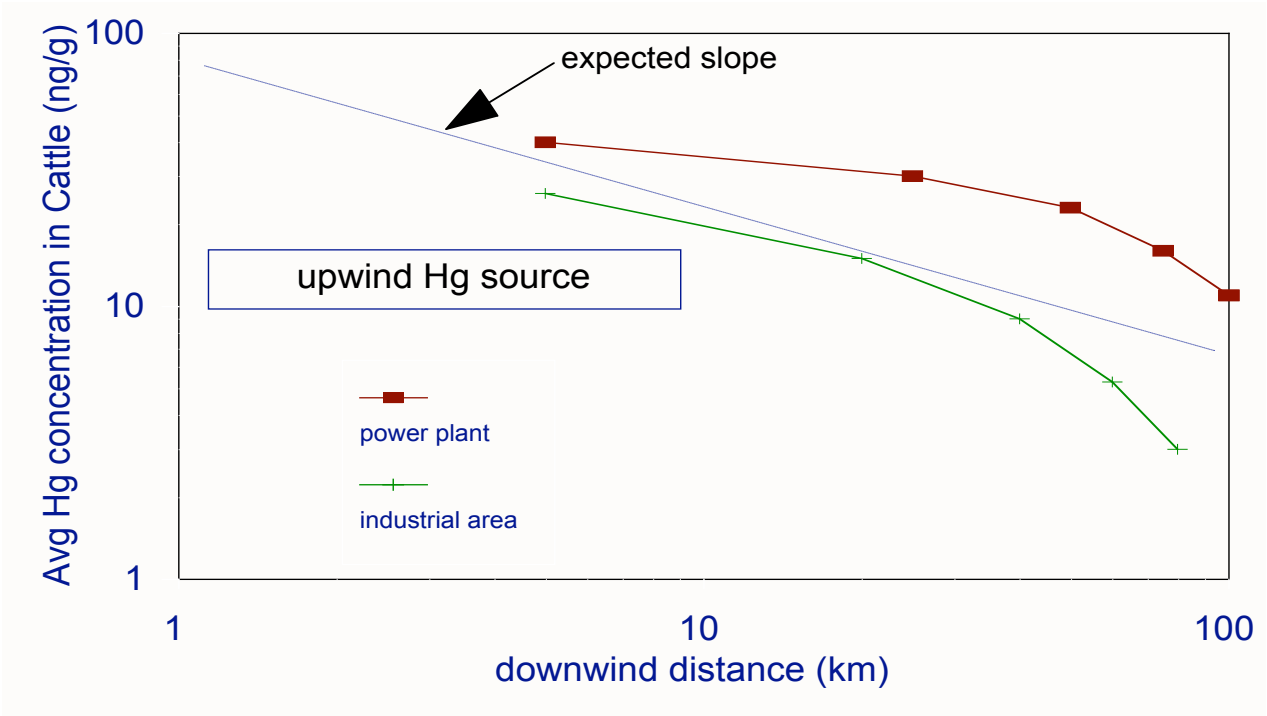

Figure 6: Measured concentration of Hg in calf kidneys as a function of distance from the source

2.3 The National Scale

2.3.1 The Relationship between Hg in Fish and Measured Wet Deposition

State-level data on Hg wet deposition from the Mercury Deposition Network (MDN) and fish Hg concentrations (EPA, 1999) were used to deduce larger scale source-receptor relationships. The deposition data were averaged over time (1997-2002) and the fish Hg data were considered by species (9 different species). A simple regression model was used in which ln(fish Hg) was regressed against ln(deposition) and dummy variables for fish species. This model assumes that all species react to deposition in the same way and provides fish Hg concentration increments relative to a referent species, in this case large-mouth bass. This approach provided 105 combinations of states and fish species (only 31 states were represented). Channel catfish, bluegills, common carp, white suckers, and yellow perch all had significantly lower Hg concentrations than bass, walleye, or northern pike (as expected). However, the effect of Hg deposition, as averaged over entire states, was significantly negative in this model ($p < 0.025$). Latitude was not an important confounder when both eastern and western states were considered. When large-mouth bass were considered alone (n=20), the relationship with deposition was positive but far from significance (log-log coefficient = 0.21).

2.3.2 Results from Global Modeling Studies

A further consideration at the national scale is provided by the recent global modeling study of (Seigneur et al., 2004), who estimated that on average only about 25-32% of the total Hg deposition to the contiguous United States is due to North American anthropogenic sources. However, this fraction of anthropogenic deposition was higher in the Northeast, where the Adirondacks receive 39% of the mercury deposited from North American anthropogenic sources. One location in New Jersey was predicted to receive 80% from these sources, but deposition at this location was dominated by Hg from incineration. These North American sources emit 200 metric tons annually, of which electric utilities comprise 53 tons, or about 26%. Assuming proportionality, the utility share of national Hg deposition would then be about 6.6-8.4%, on average, which is consistent with the various estimates based on actual measurements that were discussed above.

2.4 Discussion

The above findings lead to the overall conclusion that atmospheric deposition of Hg is affected by emissions from coal-fired power plants. However, because of the numerous assumptions required and the use of simplistic models, it is not possible to describe these relationships accurately based on these findings. Nevertheless, coal-fired plants seem to contribute less than 10% of total Hg deposition on a national scale, and the resulting effects on fish Hg would be even smaller.

Reliable quantitative understanding of the processes of mercury emissions, deposition, and translocation through the food chain remains elusive. Complex atmospheric chemistry and dispersion models are required to predict precise concentration and deposition contributions, and aquatic process models are required to predict effects on fish. However, there is uncertainty in

all of these predictions, especially with regard to the fate of RGM in coal-fired power plant plumes. Therefore, at this time, the most reliable way to understand the impacts of coal-fired power plants on Hg deposition is based on empirical data. In terms of excesses over background, local soil concentration Hg increments are around 30%-60%; sediment increments are 18-30% and wet deposition increments are around 11-12%. Soil and sediment effects are necessarily cumulative, in contrast to wet deposition. Based on the empirical finding that fish Hg is proportional to the square root of wet deposition (after controlling for water chemistry), then the contribution of coal-fired power plants to fish Hg would be about 5-6%. Local differences in water chemistry may also help explain the absence of a relationship between state-level fish concentrations and wet deposition levels; it is possible that the absence of local impacts on fish at the Kincaid power plant was related to water chemistry.

Finally, it should be noted that none of these simple analyses have accounted for possible impacts on Hg deposition from urban areas, as implied by recent findings of excess urban deposition (Nadim et al, 2001; Landis et al., 2002a). A mass-balance study of Hg deposition to Lake Michigan (Landis et al., 2002b) showed a slow rate of decrease in deposition (in terms of distance from Chicago), as did the data on power plants discussed above. This review finds no empirical evidence of bona-fide "hot spots" near coal-fired power plants, but this is not the case for chlor-alkali plants or incinerators.

3. Measuring Soil and Vegetation Concentrations of Mercury around Coal-Fired Power Plants

3.1 Introduction

Published experimental studies of deposition of mercury around point sources were reviewed in Section 2. Studies of soils, sediments, and wet deposition around coal plants typically find some evidence of enhanced deposition; however, the impact and statistical significance of the results is generally weak. The coal plant studies were conducted in the 1970's when emission rates were higher due to fewer pollution controls and the use of coals with higher Hg content. In addition, these studies did not attempt to correlate modeled deposition with measured soil concentrations.

The Hg deposition models are based on a number of assumptions and hence there is uncertainty in the predicted deposition rates. A key assumption in the models is that the mixture of reactive gaseous mercury (RGM) to elemental mercury Hg(0) is constant in the exhaust plume. However, recent experiments suggest that RGM converts to Hg(0) quickly (Edgerton et al, 2004, Laudal et al, 2004). If the hypothesis is correct, then local impact of coal-fired power plants will be greatly reduced, since Hg(0) does not deposit as quickly as RGM. In studies conducted at two sites in Georgia and one in Florida (Edgerton et al., 2004), plumes from power plants were tracked using the sulfur dioxide emitted from the stacks as a tracer. For 31 plume touchdown events, mercury speciation at the ground level monitoring station; which was 14 – 150 km away from the power plants, showed levels of RGM around 14 - 23% as compared to 53 - 75% when emitted from the stacks. Wet deposition cannot account for this reduction, as rain did not occur over the period of travel time from emission to the monitoring point (< a few hours). Dry deposition effects were also minimized by collecting samples early in the day when dry deposition is minimized. Studies of airborne mercury speciation in power plant plumes also showed a reduction in the percentage of reactive gaseous mercury. An airplane was flown through the plume from the Pleasant Prairie power plant in southern Wisconsin at distances of less than _ mile, 5 miles, and 10 miles from the stack. Measurements of the stack gas showed that releases were 67% Hg(0) and 33% RGM. Less than _ mile from the stack, the mercury speciation changed to 83% Hg(0) and by five miles approximately 88% of the mercury was in elemental form, Hg(0). This percentage of Hg(0) was maintained at the 10-mile distance. While there are uncertainties associated with measuring mercury in the plume from an aircraft, these results are consistent with the findings on the ground (Edgerton et. al, 2004) and eliminate differential deposition as the cause for the depletion of RGM.

The extant computer modeling suggests that increased local deposition will occur on a local (2 to 10 km) to regional scale (20 to 50 km) with local increases a small percentage of background deposition on the regional scale. (EPA, 1997, Sullivan et al., 2003). The amount of deposition depends upon many factors including emission rate, chemical form of mercury emitted (with reactive gaseous mercury depositing more readily than elemental mercury), other emission characteristics (stack height, exhaust temperature, etc), and meteorological conditions. Modeling suggests that wet deposition will lead to the highest deposition rates and that these will occurwithin a few km of the plant. The rates of dry deposition are are predicted to be about the same as wet deposition, but they apply to a much greater area (Sullivan et al., 2003). Therefore,

on the regional scale, dry deposition may be more important than wet. However, it is quite difficult to measure dry deposition of Hg directly.

To further understand the impacts of local deposition, soil and vegetation samples were collected around two coal-fired power plants and analyzed for mercury. One plant is a mid-size plant in the midwest that burns locally mined lignite. This site will be referred to as Plant A (for reasons of confidentiality). The second plant is the Kincaid power plant located southeast of Springfield IL that was studied in the 1970's (Anderson et al., 1977). This study combines modeling of mercury deposition patterns with soil and vegetation mercury measurements. The deposition model used emissions data, meteorological conditions, and plant data to define sample locations likely to exhibit deposition in excess of background that can be attributed to the power plant. The data are evaulated looking for revidence of 'hot-spots'. Comparisons between the data and the models were made to test the validity of the model and the input data used.

3.2 Deposition Model Paramaters

The local atmospheric transport of mercury released from the coal-fired power plants was studied to estimate the local impacts of mercury deposition. The Industrial Source Code (ISCST3) Short Term air dispersion model was used to model these processes. This code is an updated version of the computer code used by the U.S. Environmental Protection Agency to examine local deposition from combustion sources in their report to Congress in 1998 (EPA, 1997).

The basis of the ISCST3 model is the straight-line, steady-state Gaussian plume equation, which is used with some modifications to model simple point source emissions from stacks and emissions from stacks that experience the effects of aerodynamic downwash due to nearby buildings. Emission sources are categorized into four basic types of sources, i.e., point sources, volume sources, area sources, and open pit sources. Point sources were used to model the emissions from the stacks of the coal-fired power plants. ISCST3 has algorithms to simulate wet and dry deposition of mercury and depletion of the plume due to deposition. Wet deposition is modeled based on a scavenging rate that depends on the type of mercury and rainfall rate. Dry deposition is modeled based on an assumed deposition velocity. The algorithms used to in ISCST3 are described elsewhere in detail (EPA, 1995).

The ISC Short Term model accepts hourly meteorological data records to define the conditions for plume rise, transport, diffusion, and deposition. The model estimates the concentration or deposition value for each source and receptor combination for each hour of input meteorology, and calculates user-selected short-term averages. For deposition values, the dry deposition flux, the wet deposition flux, or the total deposition flux may be estimated. The total deposition flux is simply the sum of the dry and wet deposition fluxes at a particular receptor location.

Mercury emissions data from the lignite-burning power plant (Plant A) and the Powder River Basin coal-burning power plant (Kincaid) were used to represent the source terms. Meteorological data from nearby weather stations were used to simulate typical weather patterns. This approach was selected to test the consistency between model results and environmental monitoring data that suggests that measured mercury levels in environmental media and biota may be elevated in areas around stationary combustion sources that emit mercury.

Modeling deposition requires three key sets of parameters: source emissions rate, deposition parameters, and meteorological data. The next two sections describe the first two in detail. Meteorological parameters are discussed for each plant in the context of the results from deposition modeling.

3.2.1 Emissions

Two types of gaseous mercury species occur in the emissions and they behave quite differently once emitted from the stack. Elemental mercury, Hg(0), due to its high vapor pressure and low water solubility, is not expected to deposit close to the facility. In contrast, reactive gaseous mercury (RGM), Hg^{+2}, is much more soluble in water and is accommodated in rain and therefore, will deposit in greater quantities closer to the emission sources. In addition, RGM will also undergo dry deposition at a much higher rate than elemental mercury.

At the point of stack emission and during atmospheric transport, mercury can also become bound to particulate matter. This form of mercury, Hg(p), can be removed from the atmosphere by both wet deposition (precipitation scavenging) and dry deposition (gravitational settling, Brownian diffusion).

3.3.1.1 Plant A

Measured speciation data were available for Plant A. The fraction of the 2 types of mercury weighted by total emissions during the test periods is:

Hg(0) = 82.2%
Hg(+2) = 17.8%, and

The total emission from the two stacks at Plant A were 344 kg or 1.2 10^{-2} g/s.

Using the fractional release rate from the test data, the release rate for each mercury category is:

Hg(0) – 0.01 g/s
Hg(+2) – 0.002 g/s
Total - 0.012 g/s.

In addition, there was another coal-fired power plant located approximately 15 km to the southwest of Plant A. Speciation data were not available for this plant and it was assumed that it was identical to Plant A. Therefore, this plant was included in the modeling and this plant emits 267 kg of mercury per year at the following rates:

Hg(0) – 0.007 g/s
Hg(+2) – 0.0015 g/s
Total - 0.0085 g/s.

3.3.1.2 Kincaid Plant

Total mercury emissions from the Kincaid power station were 161 kg (0.0051 g/s) in 2001. Speciation data were not available, however other plants burning Powder River Basin coal generally emit 15 – 30% RGM with the remainder Hg(0). For modeling purposes, a value of 20% RGM and 80% Hg(0) was selected. Previous studies (Sullivan, et. al., 2003) showed that for less than 2% Hg(p), deposition was dominated by RGM with only a small contribution from Hg(0) over 30 km from the plant. The deposition is linearly proportional to source strength.

Therefore, errors in the estimated amount of RGM will affect the total deposition, but not the deposition pattern. Due to emission controls at the plant, less than 1% of the Hg is expected to be particulate form.

Emissions from Kincaid based on the above assumptions (g/s)
Hg(0) – 0.0041 g/s
Hg(+2) – 0.0010 g/s
Total - 0.0051 g/s

The city of Springfield also operates a small coal-fired power plant approximately 15 miles to the northwest of the Kincaid power plant. Releases from this small plant were 11 kg/yr and were also included in the modeling. Mercury release rates from the Springfield Power plant are:
Hg(0) – 0.00028 g/s
Hg(+2) – 0.00007 g/s
Total - 0.00035 g/s
Due to the separation distance and small size of this plant as compared to the Kincaid plant, modeled deposition from the small plant did not change the deposition pattern around the Kincaid plant.

3.3 Data Collection and Analysis for Plant A

Power plant A, figure 7, features two units with a total generation capacity of nearly 1,200 megawatts. The plant first started generating electricity in 1979.

Figure 7 Power Plant A

3.3.1 Deposition Modeling

Meteorological data for a five year period from the nearest airport, located about 40 miles away, were reviewed to determine wind patterns under dry and wet conditions. Under dry condtions, the prevailing winds ran along an axis from the northwest towards the southeast, Figure 8. Winds occurred regulary in each direction along this axis. Under wet conditions, winds were generally from the north and east, Figure 9. This leads to predictions of wet deposition near the plant and to the southwest. Deposition modeling of the emissions from the two power plants

(e.g. regional background was not modeled) based on the meteorological data predicted highest deposition rates within 10 km of the plant in a southwesterly direction, Figure 10. The plume

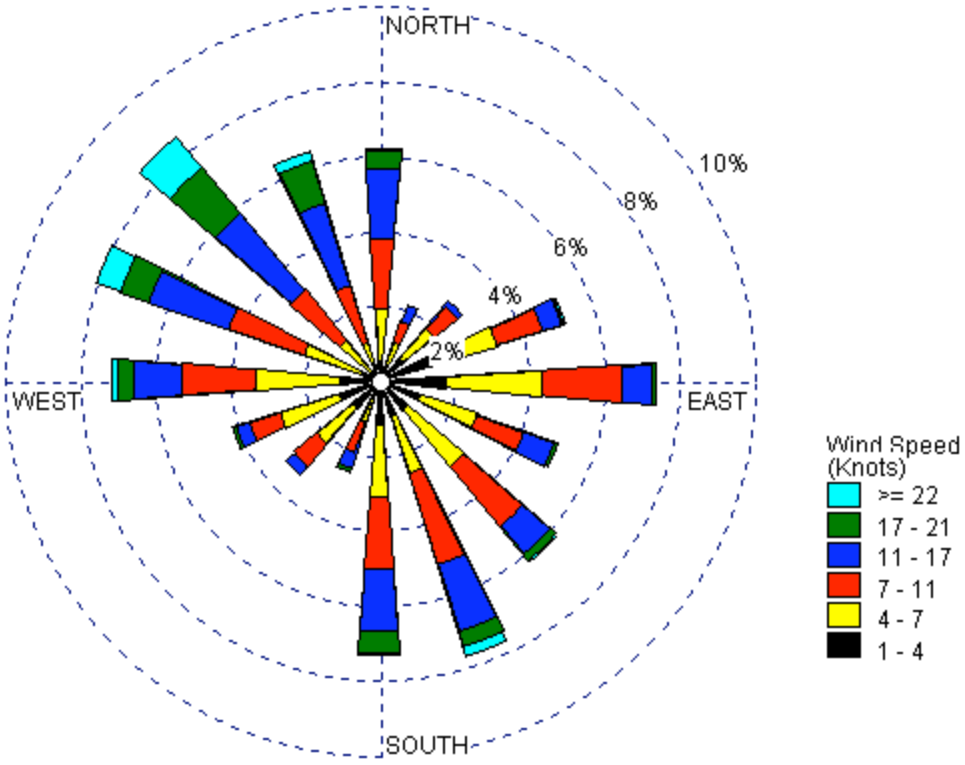

Figure 8 Direction (wind from) and intensity of wind (wind rose) used for modeling deposition near the Power Plant A.

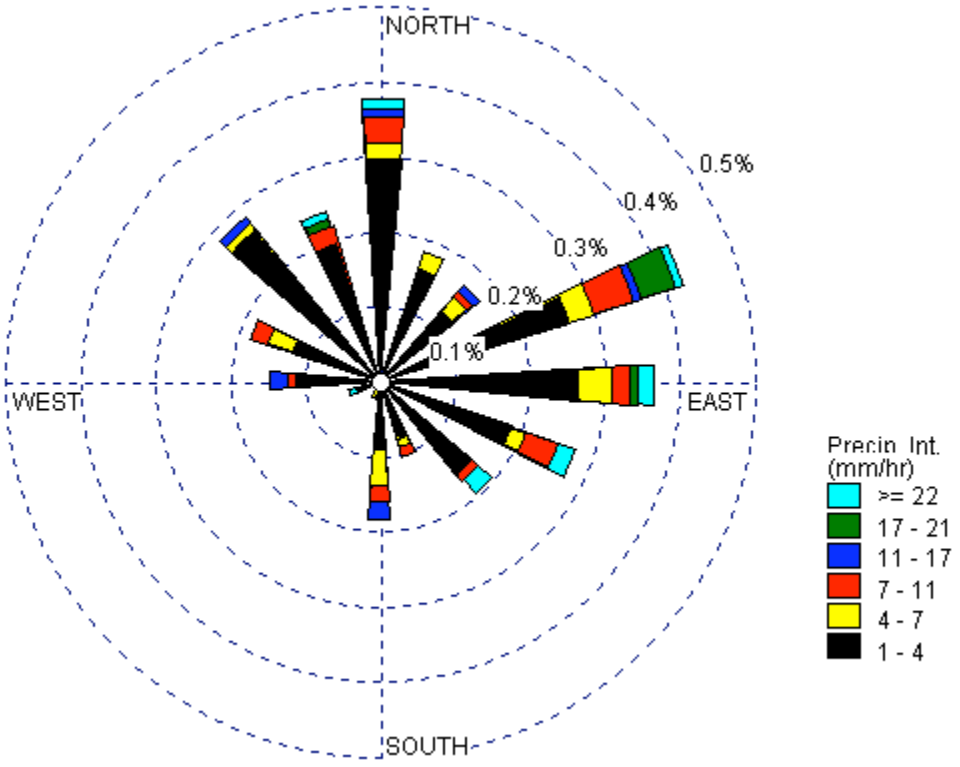

Figure 9 Precipitation intensity and direction (wind from) used for modeling deposition near the Power Plant A.

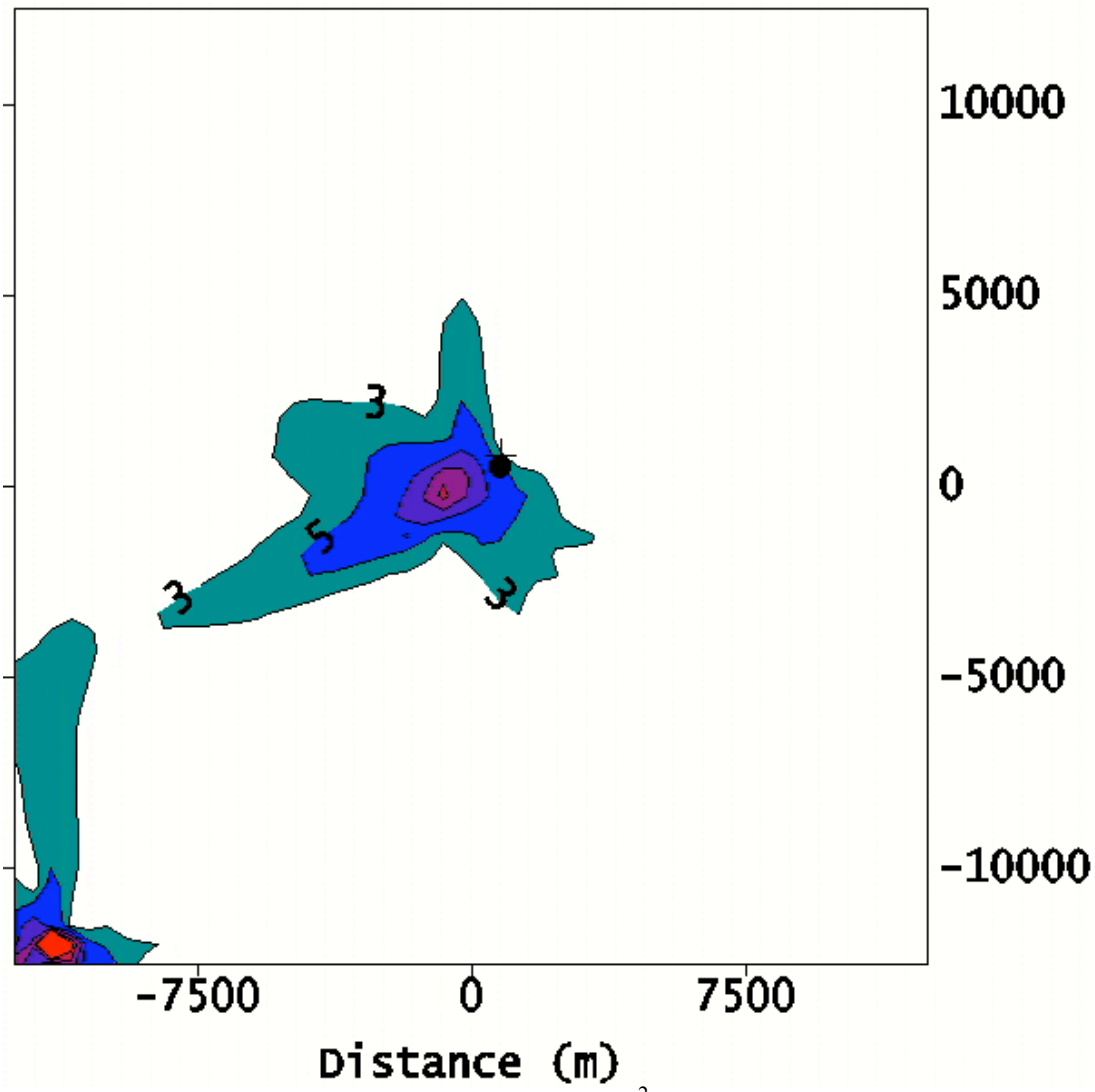

Figure 10 Modeled total Hg deposition (_g/m^2/y) pattern around plant A (located at (0,0). Second plant in the lower left corner is responsible for the deposition pattern in this region.

near (0,0) in Figure 10 is from plant A. The depositon pattern in the southwest corner (-10,000, -12,000) is from the second plant. The patterns do not show substantial overlap. Total deposition rates were 3 – 10 ug/m^2/yr. The total background deposition in this region is expected to be 10 – 20 ug/m^2/yr. Thus, the plant may produce a region of a few tens of square kilometers with depostion at 15 – 100% above background. The region with predicted deposition more than 100% above background (contour of 20 _g/m^2/y) is less than 1 km^2 in area. The region that was 15% above background deposition is less than 20 km^2 (contour of 3 _g/m^2/y). Dry depostion rates were lower than wet depostion rates and were not predicted to be a major contributor to deposition in the region. Total deposition was dominated by RGM. Over the modeled domain, deposition of Hg(0) was a small fraction of the total deposition even though 82% of the emitted mercury is in this form.

3.2.2 Sampling Design

Based on the modeled deposition analysis a soil and vegetation sampling design was selected to extend approximately 8 km to the south and west of the plant. Figure 11 shows the final sampling grid which which was modified from the general layout suggested by the modeling to

account for site-specific conditions (e.g., inaccessibility of sample locations, site activities, and changes in soil type which would alter background levels of mercury). The sampling area south and west of the plant covered an approximately square region of 64 km^2. The land surrounding the power plant was either part of an active strip mine or agricultural. Although many sampling sites were within the strip mine permit area, most of the land had been reclaimed. Strip mine personnel identified sites that had been fully reclaimed, or were at least known not to have been disturbed for at least a year. Agricultural area sampling sites were chosen because they appeared undisturbed for at least one year (i.e. had not been plowed). Many of the agricultural sites were at the crest of roadside ditches, adjacent to a plowed or mowed area.

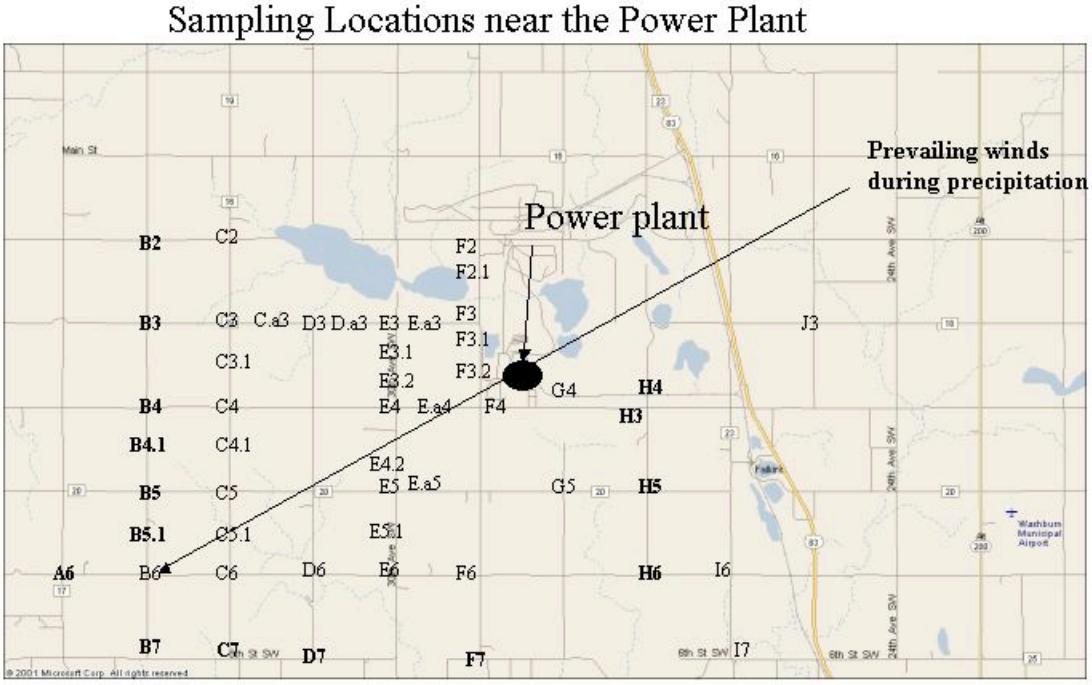

Figure 11 Soil and vegetation sample locations around the power plant.

Soil and vegetation samples were collected in November, 2003 at 54 selected sites around the coal-fired power station as shown in Figure 10. At each site, five samples were collected. Three surface samples from the top five centimeters of soil separated by approximately 3 m, one deep sample at a depth of 5 – 10 cm, and one sample of the vegetation.

Samples of approximately 100 grams weight were collected in watertight wide-mouth 250 mL plastic screw-top cups. Samples were collected using stainless steel trowels, which were rinsed with tap water and wiped dry between each use. Blind field duplicates were collected every 10^{th} sample. Samples were shipped back to Brookhaven National Laboratory (BNL) for analysis. Latitude and longitude for each sample location were identified using a GPS locator system (Garmen Etrex) with a resolution of 6 meters.

3.3.3 Mercury Analysis Methods and Quality Assurance

The soil samples were analyzed using a Direct Mercury Analyzer (DMA-80, Milestone, Inc, Monroe, CT), Figure 12. Solid samples, approximately 0.5 grams, are placed on small boats that enter the DMA. In the DMA-80, controlled heating in an oxygenated decomposition furnace liberates mercury from the solid samples. Flowing oxygen to the catalytic section of the furnace carries the decomposition products, where oxidation is completed and halogens and nitrogen/sulfur oxides are trapped. The remaining decomposition products are then carried to a gold amalgamator that selectively traps mercury. After the system is flushed, the amalgamator is rapidly heated, releasing mercury vapor, which is then carried through absorbance cells positioned in the light path of a single wavelength (253.7 nm) atomic absorption spectrophotometer. The typical working range for this method is 0.05-600 ng of mercury. Since soil samples are at most about 0.5 grams, the DMA-80 easily measures levels below 1 ppb (ng/g). DMA-80 analyses were conducted on soil samples as is. Moisture content was determined separately for all samples, and mercury concentrations were adjusted to a dry weight basis.

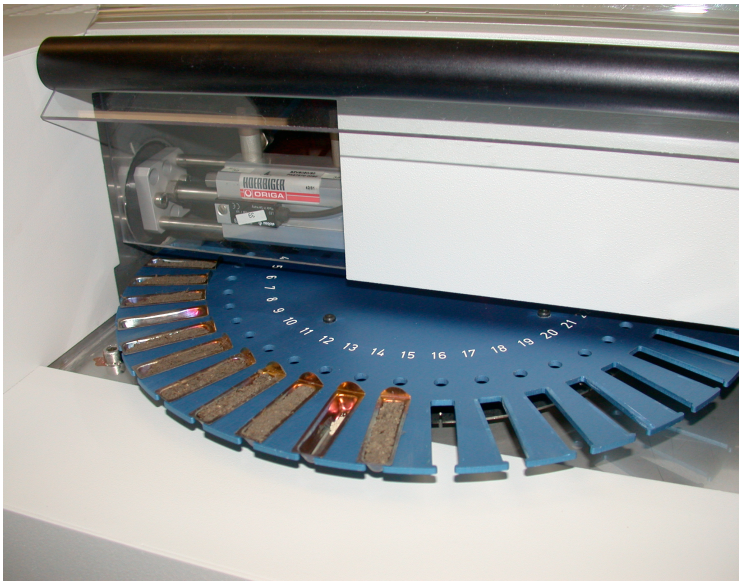

Figure 12 Direct Mercury Analyzer with soil samples

Quality assurance was evaluated through taking blind duplicates of 10% of the samples, measurement of empty sample boats in the DMA-80, and use of one of two NIST mercury standards (SRM 2709 and SRM 2710) at every 10th measurement. The NIST soil standard SRM 2709 San Joaquin soil) was used for soil measurements. It has a mercury level of 1380 +/- 80 ng/g. Figure 13 shows the results of measurements of the San Joaquin QA standards taken during one set of soil samples. The NIST standard SRM 2710, peach leaves, was used for vegetation samples. It has a mercury level of 30 +/- 5 ng/g. Empty boat samples averaged 1.4 ng/g and were all less than the minimum soil sample (10 ng/g). Blind duplicates were statistically similar to the similar soil samples.

Each sample was measured in triplicate to examine the homogeneity of the sample. The range of the mercury levels in the three samples averaged +/- 12.5% of the average of the three samples.

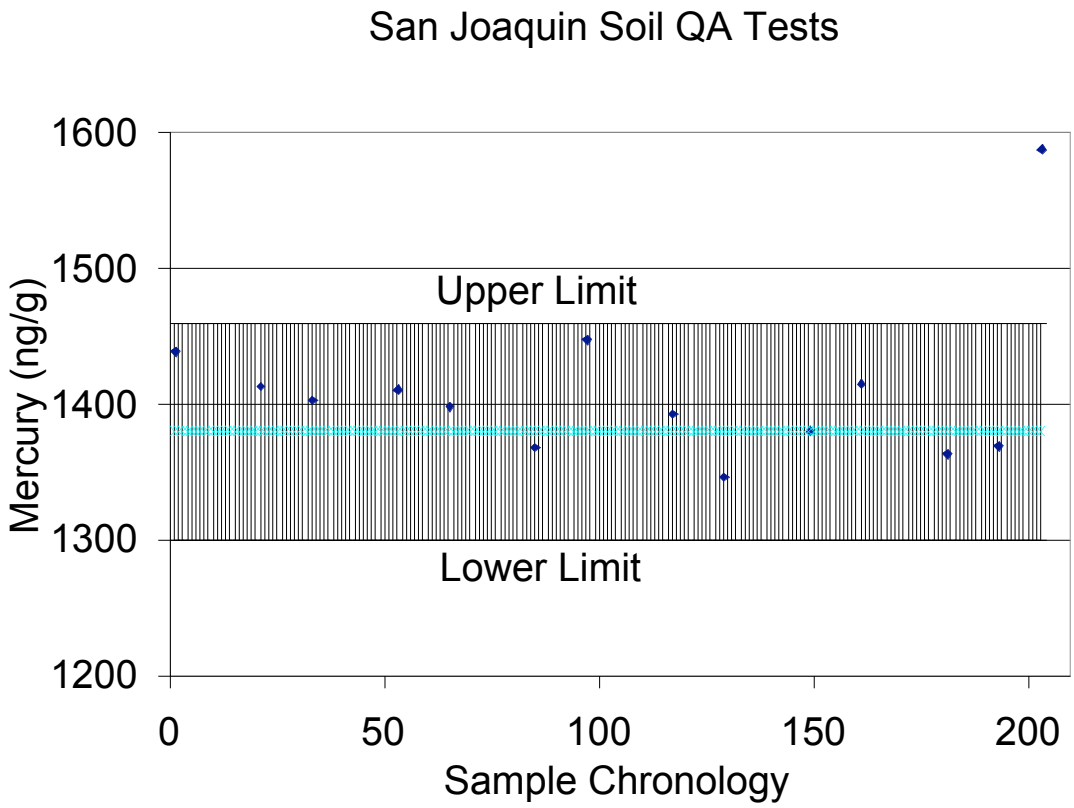

Figure 13 NIST Standard San Joaquin soil measurements.

3.3.4 Data Analysis and Interpretation

3.3.4.1 Soil Data

At each sample location, the three surface soil samples were averaged to give a composite. Figure 14 provides the distribution of soil Hg data as a function of probability. Analysis of the data shows that they are log normally distributed, as would be expected for soil samples (Tack, 2005). At the fifty-four locations the average value was 27.6 ng/g (dry weight basis), with a standard deviation of 6.9. The minimum value was 11.6 ng/g and the maximum value was 55.4 ng/g. All of the data were within approximately a factor of 2 of the average value thereby suggesting that 'hot-spots' were not found in the soil near the plant, in contrast to the findings for chlor-alkali plant discussed in Section 2. For example, the four highest values (> 36 ng/g) in Figure 14 look like they might be outliers, but only two are contiguous and none fall into the patterns predicted by modeling. Averaging these values with their nearest neighbors, to simulate what might occur in a sizeable water body, reduces their magnitudes considerably: to the southeast, 31 ng/g; to the northeast, 29.9 ng/g,; to the south, 29 ng/g. These values are from 5-13% above the median of all samples (27.5 ng/g), and do not constitute "hot-spots" in the conventional sense.

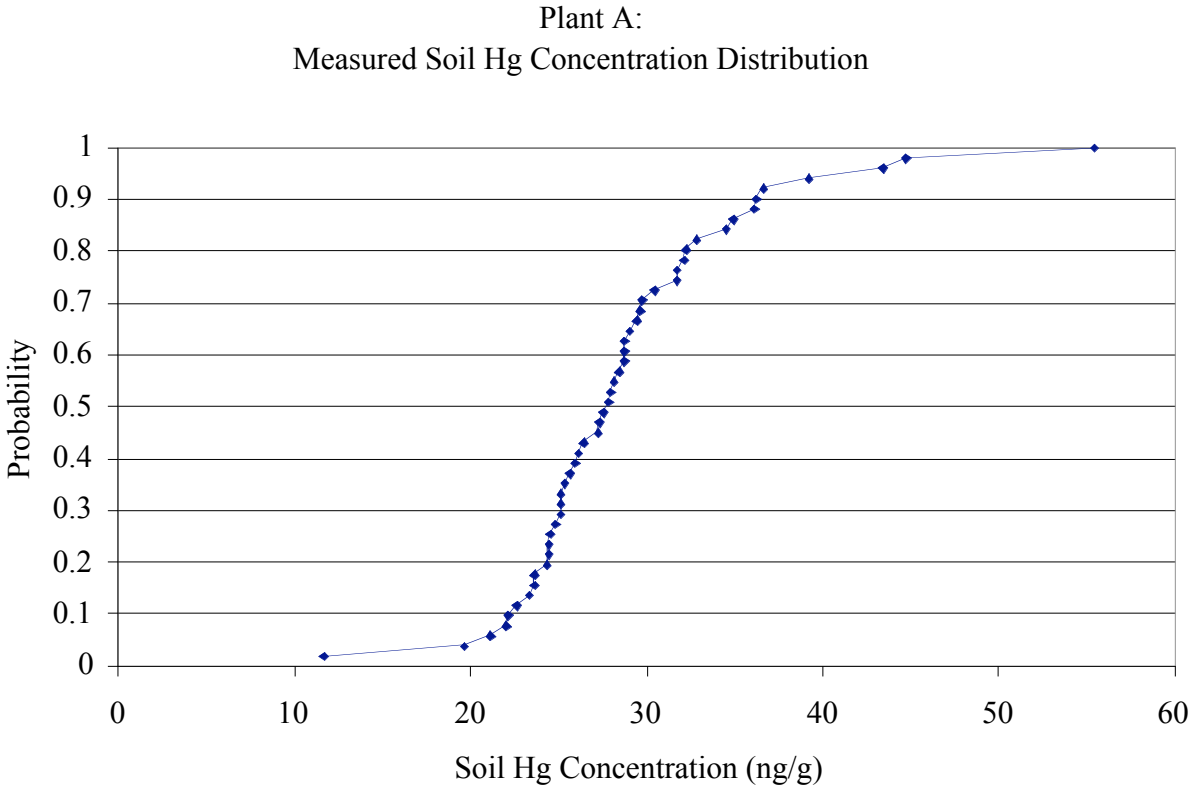

Figure 14 Distribution of soil Hg concentration in 54 samples from around Plant A.

Comparison between the predicted deposition versus measured mercury concentrations in the soil was accomplished by overlaying the deposition map over the sampling map with sample results color-coded by measured concentration. Figure 15 is the graphical representation of the analysis. Soil concentrations were binned into three approximately equal size groups containing 17 or 18 samples. Sample locations with symbols representing measured mercury levels represent the measured data. The range of the groups was 11.4 – 25.1, 25.1 – 29 and 29 – 55.4 ng/g. The tight grouping of the middle group around the average shows that 1/3 of the soil Hg measurements were in this narrow range. Predicted regions of enhanced deposition are covered by the filled contours with red representing 5 ug/m^2/yr and blue representing 3 ug/m^2/yr.

The model predicts the incremental rate of deposition due to the coal plant emissions, while the measured data are soil concentrations, reflecting the effects of cumulative deposition, both local and regional, and eh natural constituents of the soil. Therefore, a direct comparison between the modeled results and the measurements is not possible. However, if excess deposition were occurring in a region, it is expected that this would be reflected by higher soil concentrations. Figure 15 shows that the patterns of modeled deposition and measured data do not match. The measured soil data suggest that the main finger of the plume is slightly south of the area predicted by modeling. Also, the measured data shows a fair degree of scatter, as expected, in contrast to the smoothly varying deposition pattern. To evaluate if there was a match between the data and the model statistically, both were ordered from high to low and a rank correlation between the soil and predicted deposition was performed. The Spearman rank correlation coefficient was -0.02 indicating no correlation between the two.

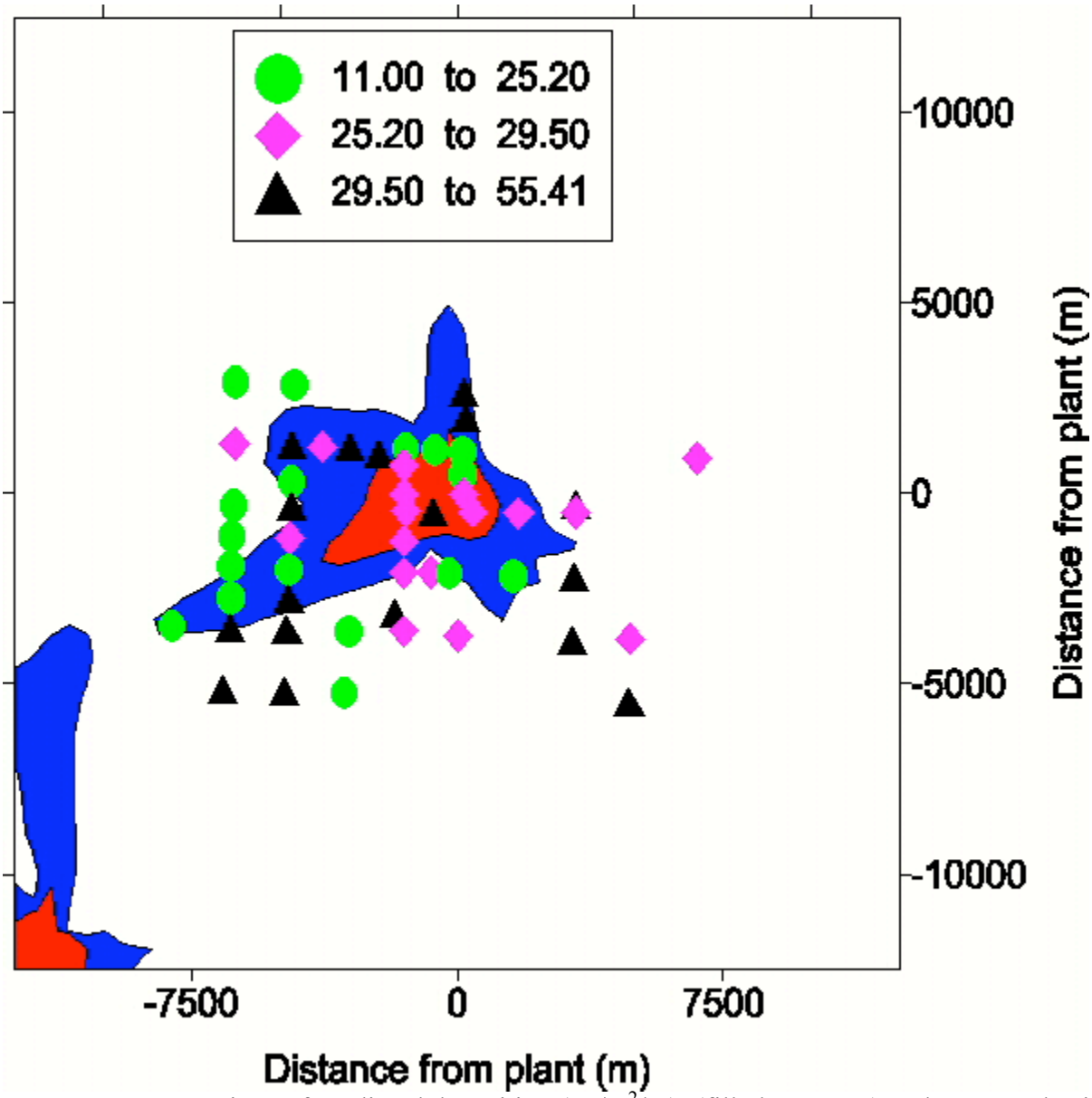

Figure 15 Comparison of predicted deposition (ug/m^2/yr) (filled contours) and measured soil Hg concentrations (ng/g) (symbols)

A second approach to determining if excess deposition was occurring was to compare the concentrations of surface soil (0 – 5 cm) and subsurface soil (5 – 10 cm) collected at each sampling location. The subsurface samples showed similar values and characteristics as the surface samples. The average value was 28.2 ng/g with a range of 10 – 49 ng/g for the subsurface samples. Figure 16 shows a comparison of surface and subsurface soil Hg levels showing the similarities. The slope is essentially 1:1 and the Spearman Rank correlation coefficient is 0.77 indicating a high degree of correlation. Therefore, once again, there is no evidence that local deposition increased the surface Hg soil concentrations relative to the subsurface soil.

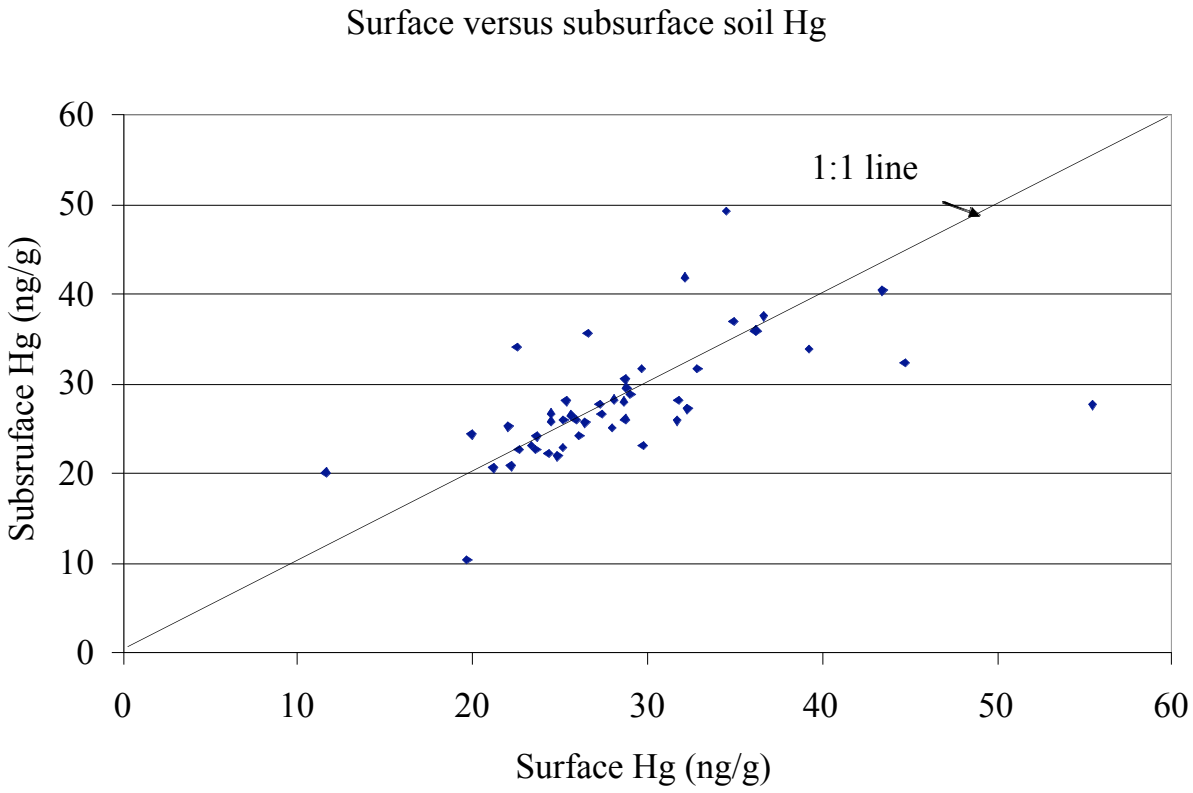

Figure 16 Comparison of surface and subsurface soil Hg concentrations.

Although there is no strong evidence of excess deposition near the plant, estimates of the potential deposition were performed for the 8 km (5 mile) quadrant to the southwest of the plant where data were collected. The following assumptions were used to estimate excess deposition.

Case a) the difference between the average surface and average subsurface soil values. This value is 0.6 ng/g.
Case b) the difference between the average surface concentration and the average of the lowest 1/3 surface concentrations. This assumes that the pre-operational background was the average of the lowest 1/3 of the samples. This 'background' average is 22.4 ng/g and the average of all soils was 28.8 ng/g.
Case c) the predicted modeled yearly deposition generated by ISTCST (408 g/yr) multiplied by the operational period (23 years).

Using a soil density of 1.5 g/cm^3 and limiting the excess deposition to the top 5 cm give a total volume of soil of 2.55 10^6 m^3 and a soil mass of 3.8 10^{12} g over the study area. Assuming that current mercury emission rates and speciation distribution apply over the 23-year operating period of the plant, Table 2 presents the estimated amount deposited, the % of RGM emissions, and the % of total emissions that this represents. The table shows that for any scenario, less than 2% of the RGM (less than 0.5%) of the total mercury emissions from the plant deposited within this 8 km square quadrant to the southwest of the plant. Assuming similar deposition in the other 3 quadrants suggests that less than 7% of the RGM emitted deposits within 8 km (5 miles of the plant). This analysis also supports the contention that although mercury levels may be slightly

elevated near the plant, a 'hot spot' large enough to affect water quality and fish Hg does not occur in the area sampled near the plant.

Table 2 Estimates of mass deposition and fraction of mercury emitted that was deposited for Plant A

Case	Total Mass Deposited (g) over 23 year plant life	% of RGM	% of Hg(total)
Surface vs. Subsurface Avg	2200	0.16	0.023
Lowest 1/3 avg as background	24500	1.7	0.3
ISCST Model	9400	0.66	0.12

3.3.4.2 Vegetation Concentrations

One vegetation sample was collected at each sampling location. The vegetation samples are a measure of mercury deposition over the current growing season. Vegetation mercury levels are known to be influenced by both wet and dry deposition of mercury. In sampling, every attempt was made to collect the same type of vegetation, grass, from each location. This was not always possible, but all samples are grasses. The samples were analyzed in duplicate and the average value was taken as a measure of the Hg content for samples that were low in mercury and provided similar readings in both measures. Many samples showed high levels of Hg (> 1ppm) and wide variability in the measured value. These samples were analyzed between 3 and 7 times to improve the accuracy of the results. The statistics from the average at each location are in Table 3.

Table 3 Statistical parameters for Vegetation Samples at Plant A.

	Value (ng/g)
Min	10.7
Max	653.3
Median	31.8
Average	96.7 (Mean of the logs 1.7)
Standard Deviation	143.5 (Standard Deviation of the logs 0.47)

Unlike the soil samples, the vegetation samples did show regions of elevated Hg concentration. Defining a high Hg concentration as any average value that is 3 times above the median value for all samples shows that 12 of 49 samples have 'high' concentrations. Individual samples were in excess of 1000 ng/g. Although, there was wide variation in the measured mercury value for the vegetation with 'high' levels of mercury (>50 ng/g), these samples were consistently high in all of the samples.

Figure 16 presents the measured mercury concentration in vegetation and the predicted deposition. In Figure 16, the blue contour represents excess deposition of 3 ug/m^2/yr and the red represents 5 ug/m^2/yr. Soil vegetation samples were divided into four groups representing the range from 10.7 – 22.9; 22.9-31.5, 31.5 -84.4; and 84.4 – 691 ng/g. Each range had 12 or 13 members and therefore, they represent _ of all samples. Approximately _ of the samples were

within 400 m of the plant. In Figure 16, the highest values are near the plant, primarily to the north and west. This is the primary direction of wind flow during dry deposition conditions. However, deposition modeling would predict that the peak deposition rates would occur further from the plant due to the stack height and buoyancy effects. All of the values in the top _ of the distribution are within 4000 m of the plant with the exception of one value located approximately 6500 m east of the plant. This site was selected to be out of the predicted deposition pattern and was hoped to be representative of background. Clearly, it is not. Although the frequency distribution of the entire set of samples appears to be reasonably continuous, the spatial distribution is not. Most of the very high values are located adjacent to a coal transporter that was previously used to move coal from the mine to the plant.

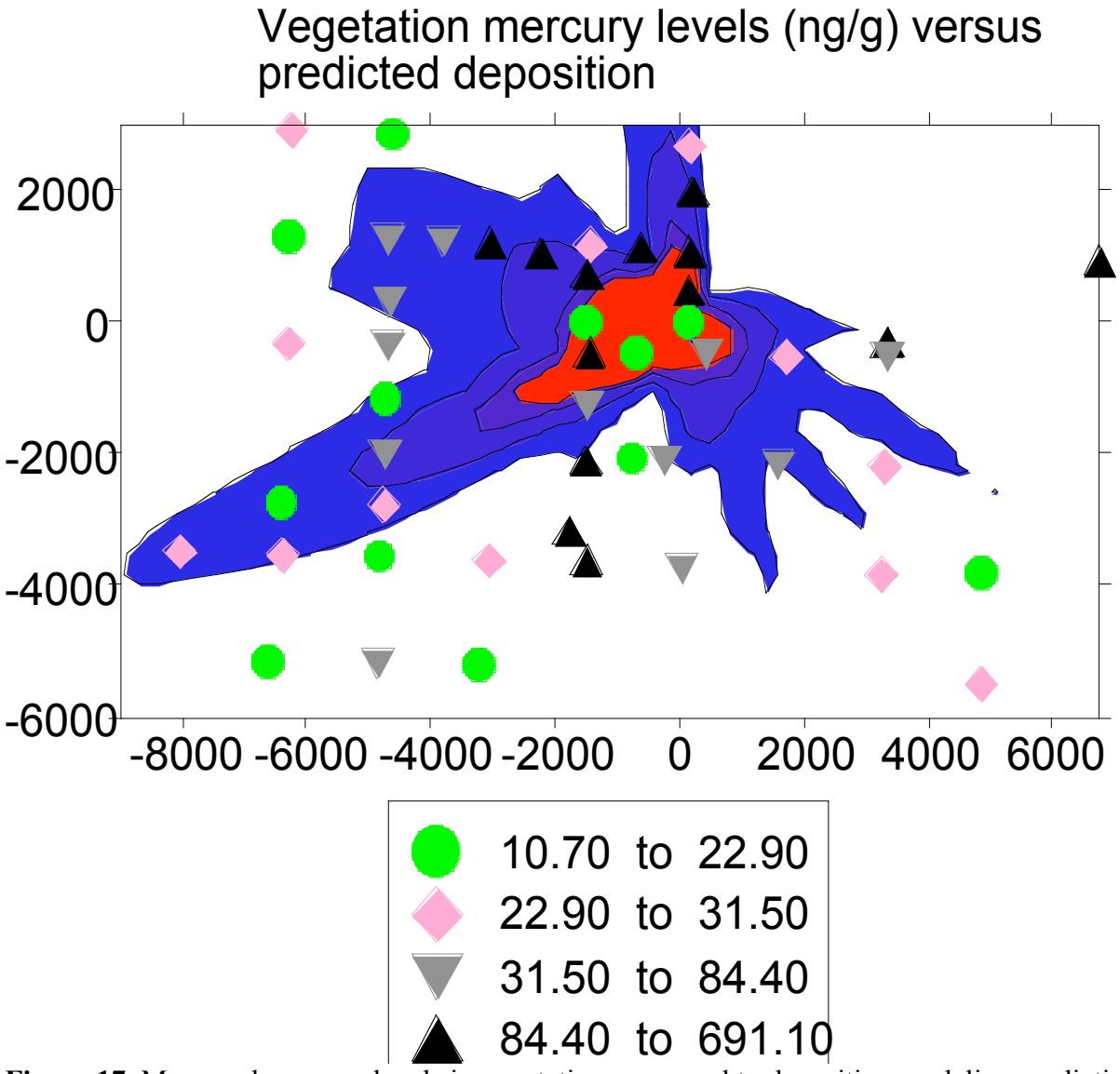

Figure 17 Measured mercury levels in vegetation compared to deposition modeling predictions.

Table 4 presents the average concentration as a function of distance from the plant for the vegetation and soil samples. The vegetation samples show a peak between 1500 – 3000 m from the plant and a decline after that point. The soil samples do not show any significant trend with distance. This data suggests that localized Hg hot spots were seen in the area, but they were not

clearly associated with the emissions from the power plant. Possibilities for the high Hg concentrations in vegetation include fugitive emissions (e.g. dust from coal handling, etc.) and proximity to roads. To resolve this issue would require additional sampling and analysis for other elements that are found in coal plant emissions.

Table 4 Average vegetation and soil concentrations as a function of distance from the plant.

Distance	# of vegetation samples	Average Hg concentration in vegetation (ng/g)	# of soil samples	Average Hg concentration in soil (ng/g)
160 – 1500 m	5	37.4	5	28.8
1500 – 3000 m	10	179.4	10	27.0
3000 – 4500 m	8	153.4	8	29.9
4500 – 6000 m	10	106.8	10	31.1
6000 –7500 m	13	29.2	13	24.9
7500 – 9000 m	6	23.2	6	30.0

3.4 Data Collection and Analysis for the Kincaid Plant

The Kincaid plant was selected because it has been studied in the 1970's (Anderson, 1977) for increases in mercury content in the soil, sediment, air, and fish. In addition, tracer studies were performed on the emissions from this plant and measured concentrations were compared with predictions by the ISCST computer code (Cox, et. al., 1986). This provides confidence that the ISCST models reasonably represent the plume at this site. The Kincaid plant began operation in 1967 and has two 575 MW boilers each with a 187 m exhaust stack.

Figure 18 Kincaid Power Plant

In addition to the Kincaid Plant, the city of Springfield operates a small plant that emits less than 9 kg (20 lbs) of mercury per year. This small plant is approximately 15 miles northwest of the Kincaid plant. Both plants were simulated in the deposition modeling.

3.4.1 Deposition Modeling

Meteorological data from the Springfield airport, located about 20 miles away, were reviewed for a five-year period to determine wind patterns under dry and wet conditions. Under dry condtions, the prevailing winds were primarily from the south, Figure 19. Winds occurred from the south 20% of the time. No other direction had winds more than 10% of the time. Winds rarely came from the Northeast. Under wet conditions, winds were generally from the south and north, Figure 20. However, precipitation also fell frequently when winds were out of the east. This leads to predictions of wet deposition near the plant and along the north south axis.

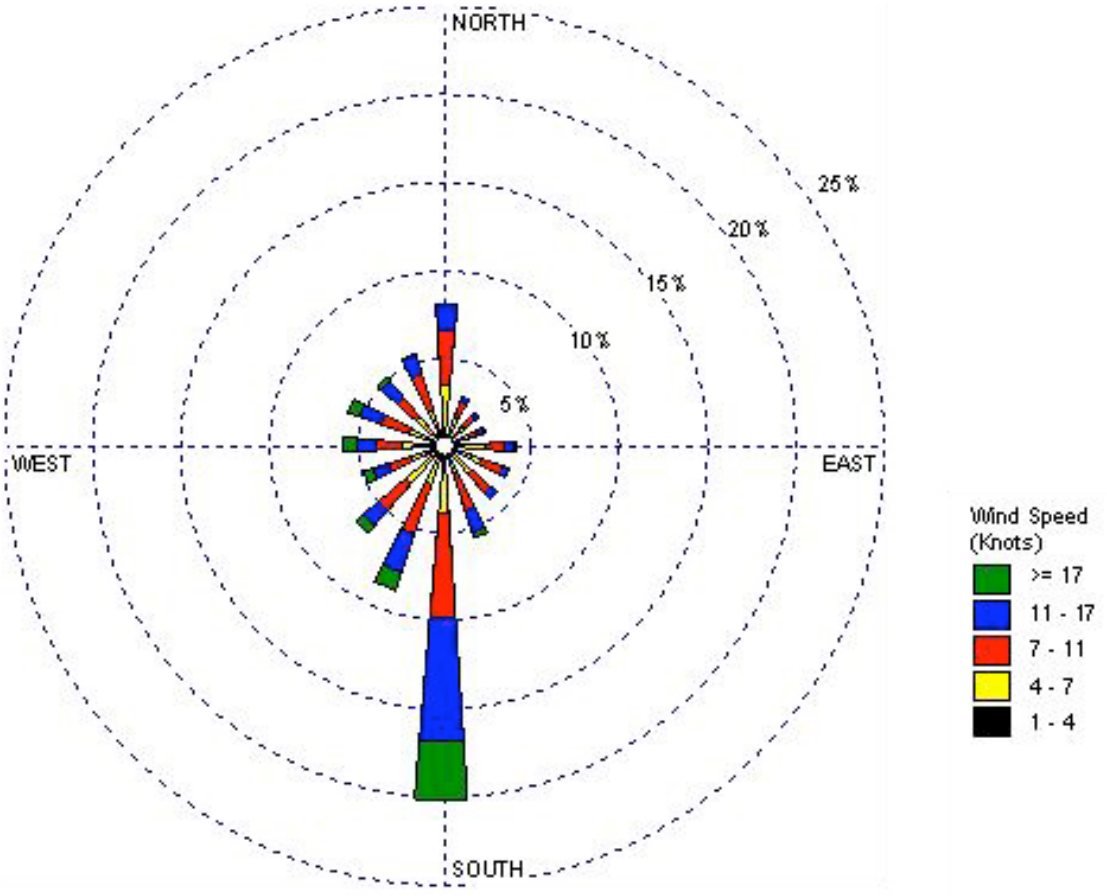

Figure 19 Wind rose for Springfield, IL near the Kincaid Power plant.

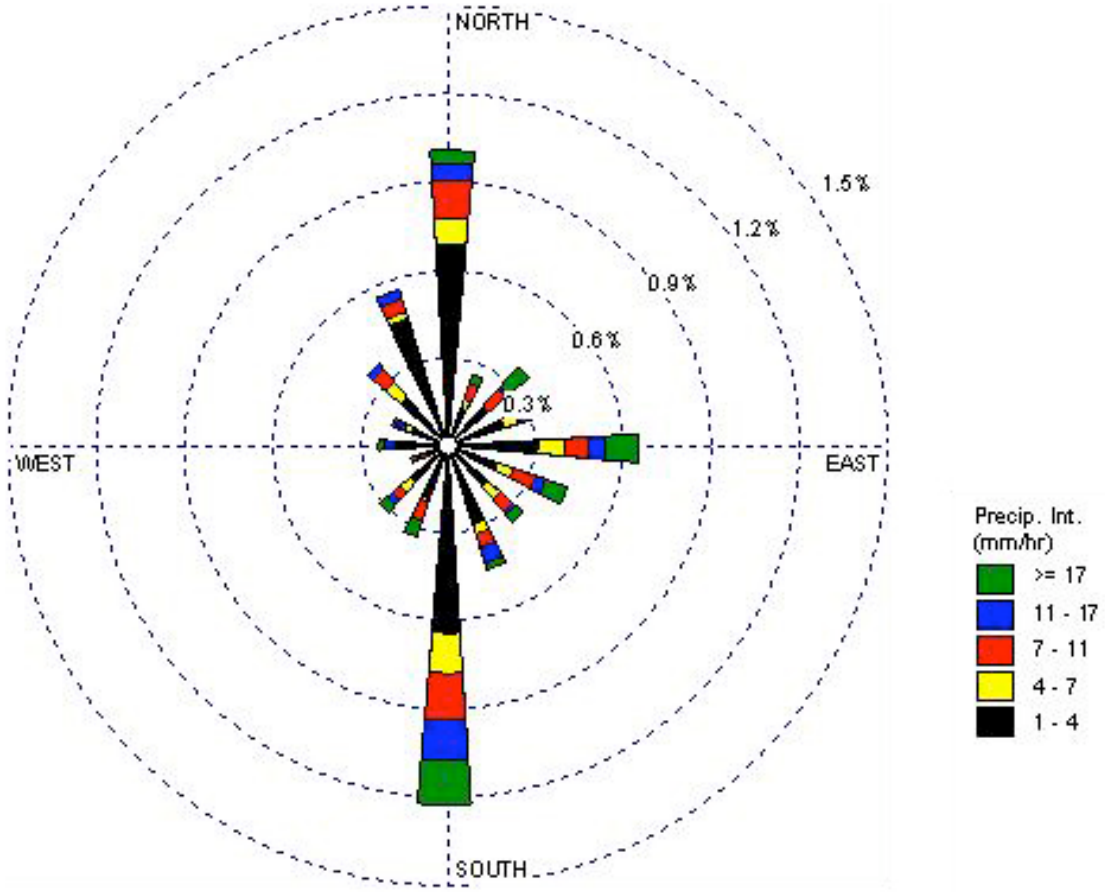

Figure 20 Rain rose for Springfield IL near the Kincaid Power Plant

Deposition modeling of the emissions from the two power plants based on the meteorological data considered both dry and wet depositon; however, regional background was not modeled. Simulations were performed for the 5 year period from 1986 – 1990. Ths period included annual rainfall ranging from 24 to 46 inches and was representative of the range of condtions experienced in the region. From the results annual deposition rates were calculated for both dry and wet deposition. Dry deposition is predicted to peak approximatley 18000 m north of the plant at a rate of 0.6 ug/m^2/yr, Figure 21. The highest wet deposition rates were predicted within 10 km of the plant in a northerly direction, Figure 22. The predicted peak deposition rate was 18 ug/m^2/yr at 1000 m north of the plant (this is the first calculated point). The deposition plume remains above 5 ug/m^2/yr for approximately 2 or 3 km around the plant and above 1 ug/m^2/yr for 7 or 8 km. For comparison, in the period of 1999 – 2003, wet deposition (due to all sources including the regional background) averaged 9.3 ug/m^2/yr at the mercury deposition network (MDN) site in Bondville, IL which is approximatley 100 km west of the plant. Dry deposition is not well characterized but is expected to be approximately the same as wet depostion. If this is true, at the location of the predicted peak deposition near Kincaid, total Hg deposition would be approximately twice background. Depostion rates resulting from Kincaid emissions would add less than 10% of background after 8 km.

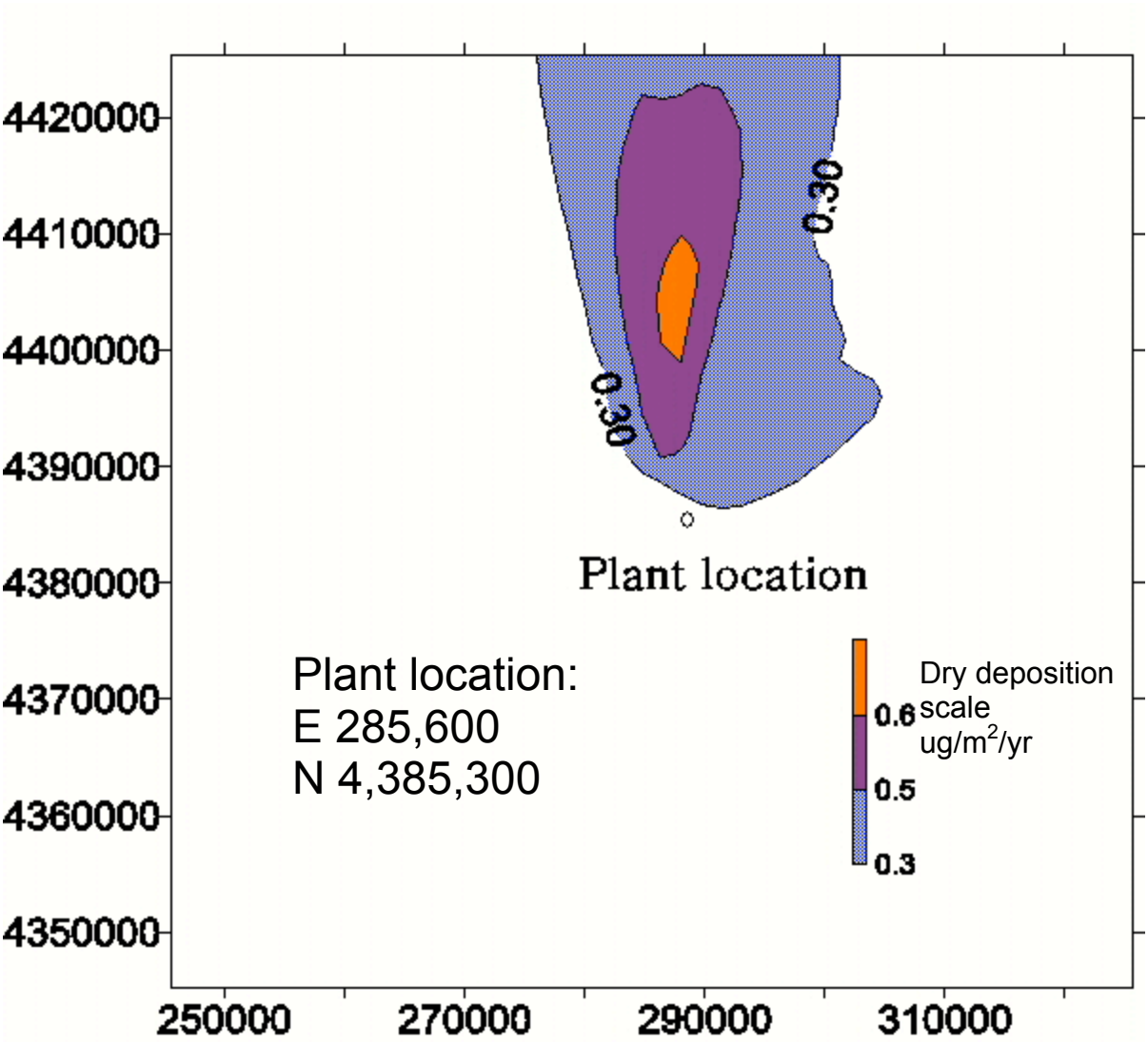

Figure 21 Predicted dry deposition resulting from Hg emissions from the Kincaid Power Plant.

Figure 22 also illustrates the impacts of dry deposition on the overall depositon pattern. The colored contours represent the wet deposition at levels of 0.5, 1, 5, and 10 ug/m^2/yr. The dotted contour line represents the total deposition (wet and dry) and shows the extension of this region to the north of the plant due to dry deposition. At deposition rates above 1 ug/m^2/yr, dry deposition does not impact the contours because the predicted dry depostion rate in this region is much less than 1 ug/m^2/yr.

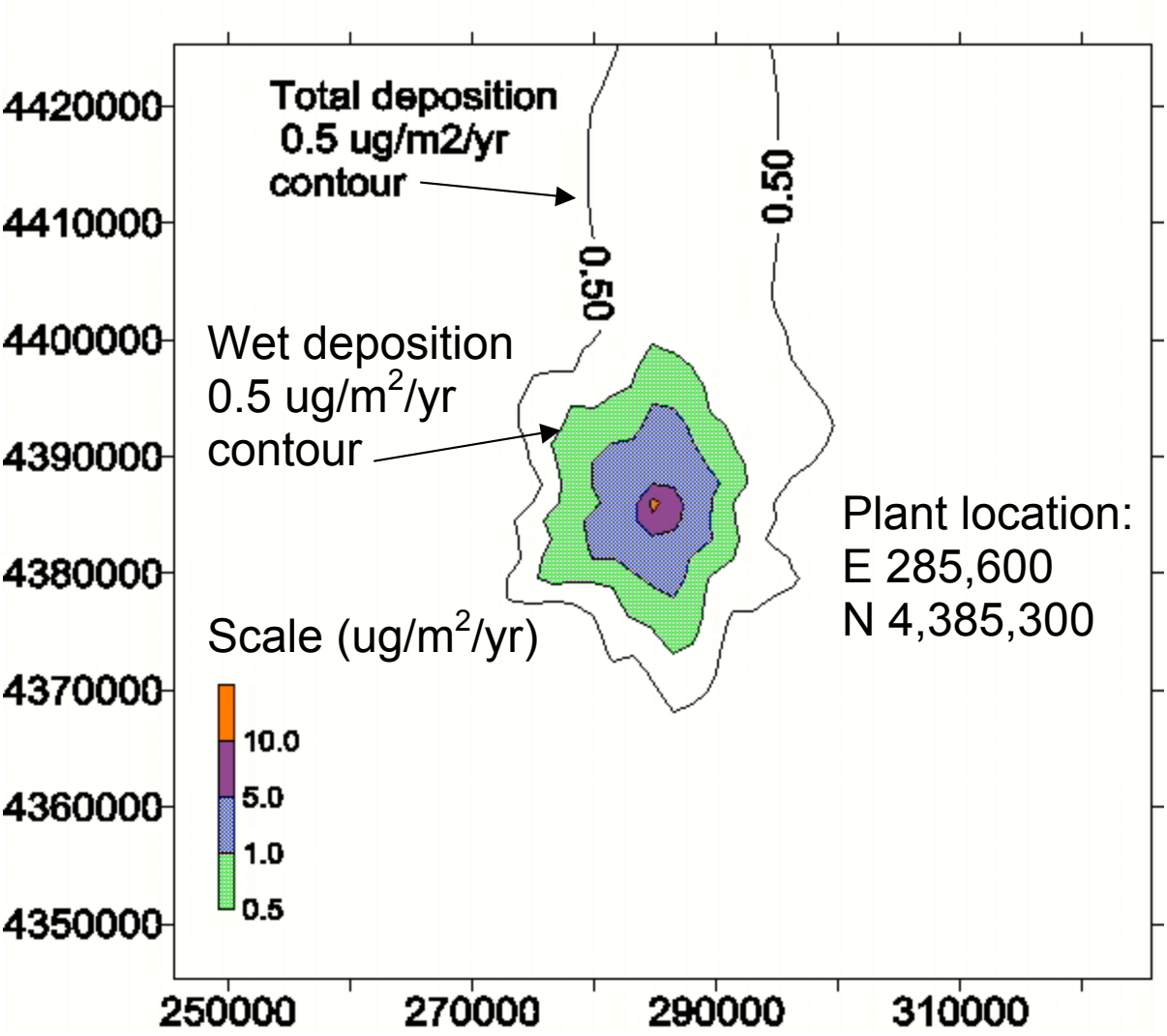

Figure 22 Predicted mercury deposition wet and total deposition contours from emissions from the Kincaid power plant.

3.4.2 Sampling Design

Even though the deposition modeling predicted higher deposition to the north of the plant, due to the absence of a strong signal in the soil mercury levels at Plant A, it was decided to sample all directions uniformly around the plant. A sample grid centered around the plant and extending approximately 8 km in every direction was developed. Samples were spaced approximately 1600 m (1 mile) apart. A total of 123 sample locations were selected. In addition, 8 additional sites were chosen with the intent of defining background. These sites were from 17.5 – 38 km from the power plant. At each sampling site, 3 surface samples (0 – 5 cm), 1 subsurface sample (5 – 10 cm) and 1 vegetation sample were collected in July, 2004. With 5 samples at each of the 131 locations, this results in 655 mercury analyses for the complete set. Samples were collected approximately 15 - 20 feet from the road edge in regions that were not plowed for farming.

Eight sample locations were less than 10 feet from the road due to space limitations. The sampling network near the plant is shown in Figure 23.

The region around the plant is mostly open farmland. Lake Sangchris, a state park, is located directly to the north and slightly east of the plant in the region of highest predicted deposition. Lack of access roads in this region prevented higher density sampling. Figure 24 shows the selected background sites for sampling.

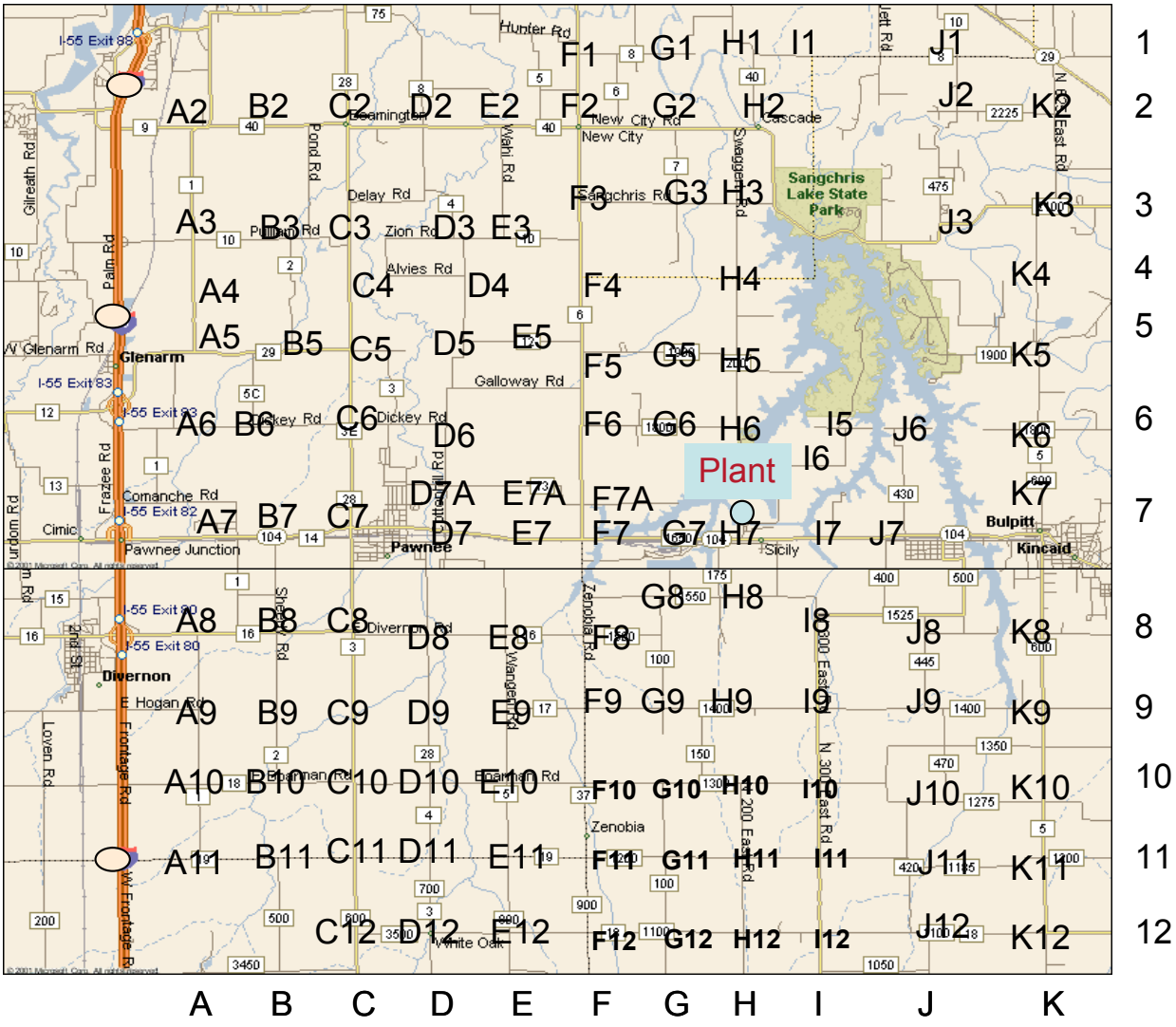

Figure 23 Sample grid around the Kincaid Power Plant.

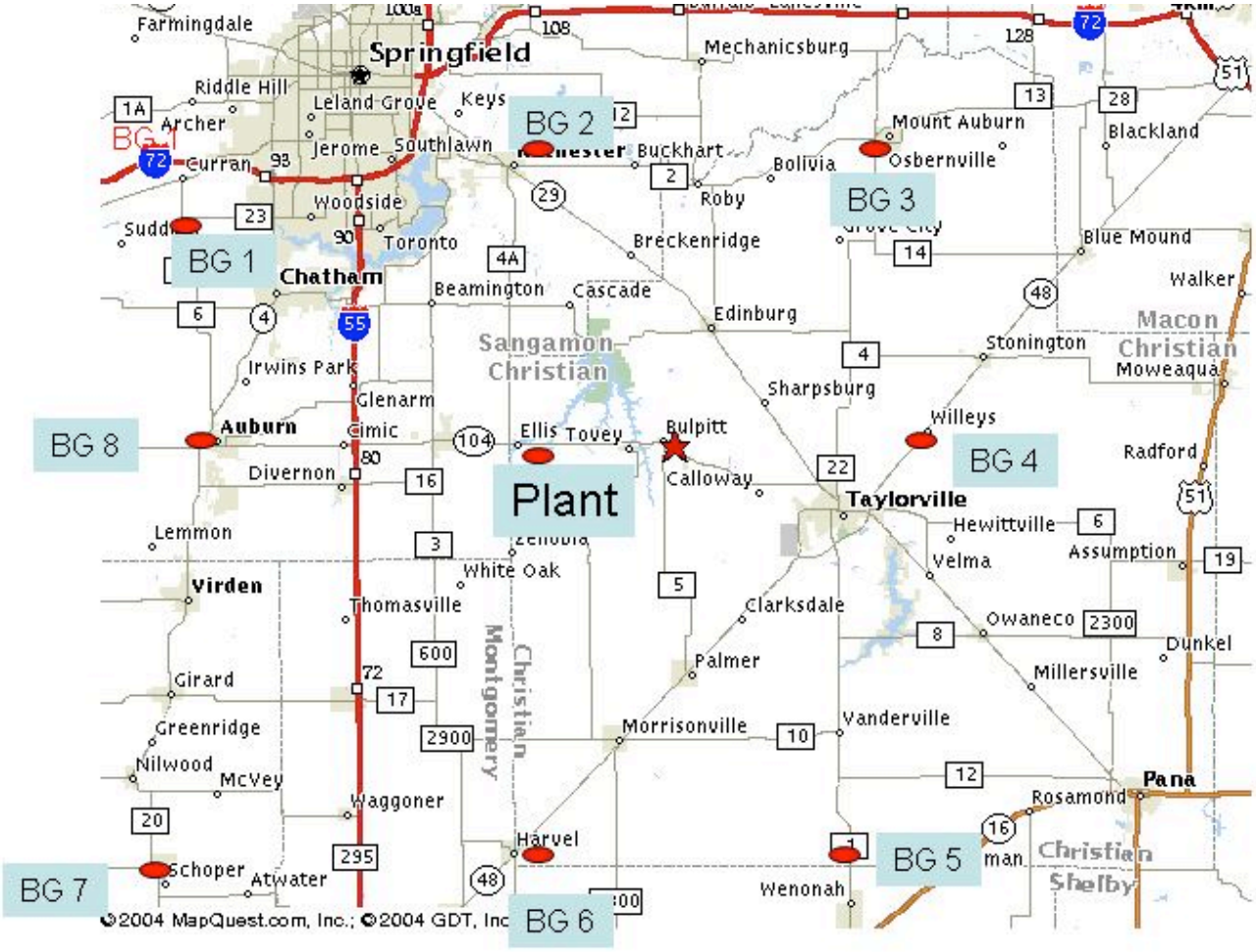

Figure 24 Background sample locations.

3.4.3 Mercury Analysis Methods and Quality Assurance

The soil samples were analyzed using a Direct Mercury Analyzer (DMA-80, Milestone, Inc, Monroe, CT), Figure 12. Moisture content was determined separately for all samples. Quality assurance was evaluated through taking blind duplicates of 10% of the samples, measurement of empty sample boats in the DMA-80, and use of one of two NIST mercury standards (SRM 2709 and SRM 2710) at every 10th measurement. Blind duplicates were statistically similar to the similar soil samples. Each sample was measured in duplicate to examine the homogeneity of the sample. The average from the two rounds of sampling differed by 6% indicating that the measurements were reproducible.

3.4.4 Data Analysis and Interpretation

3.4.4.1 Soil Data

A total of six values (3 locations and duplicate measures at each location) were used to estimate the average at each sample location. At the 124 locations around the plant, the average value was 32 ng/g (dry weight basis), with a standard deviation of 17.7 (GSD =1.34). The median value was 25.9 ng/g. The minimum value averaged over the six measurements was 16.9 ng/g and the maximum value was 155.6 ng/g. The minimum value for any single sample was 12 ng/g

and the maximum for any single sample was 218 ng/g. The data showed a much wider distribution than for Plant A with a few values that were very high above the mean. Figure 25 provides a normal Q-Q probability plot for the log of soil mercury versus theoretical quantile. The plot shows the data and the log normal distribution that would occur if the mean of the log soil mercury and standard deviation were used to estimate concentrations. If these two lines are very similar, it suggests that the data follow a log normal distribution. It is clear that this is not the case with this data set. The data are skewed towards higher values above a theoretical quantile of 1.0. This could imply enhanced deposition, or it could imply differences due to different soils.

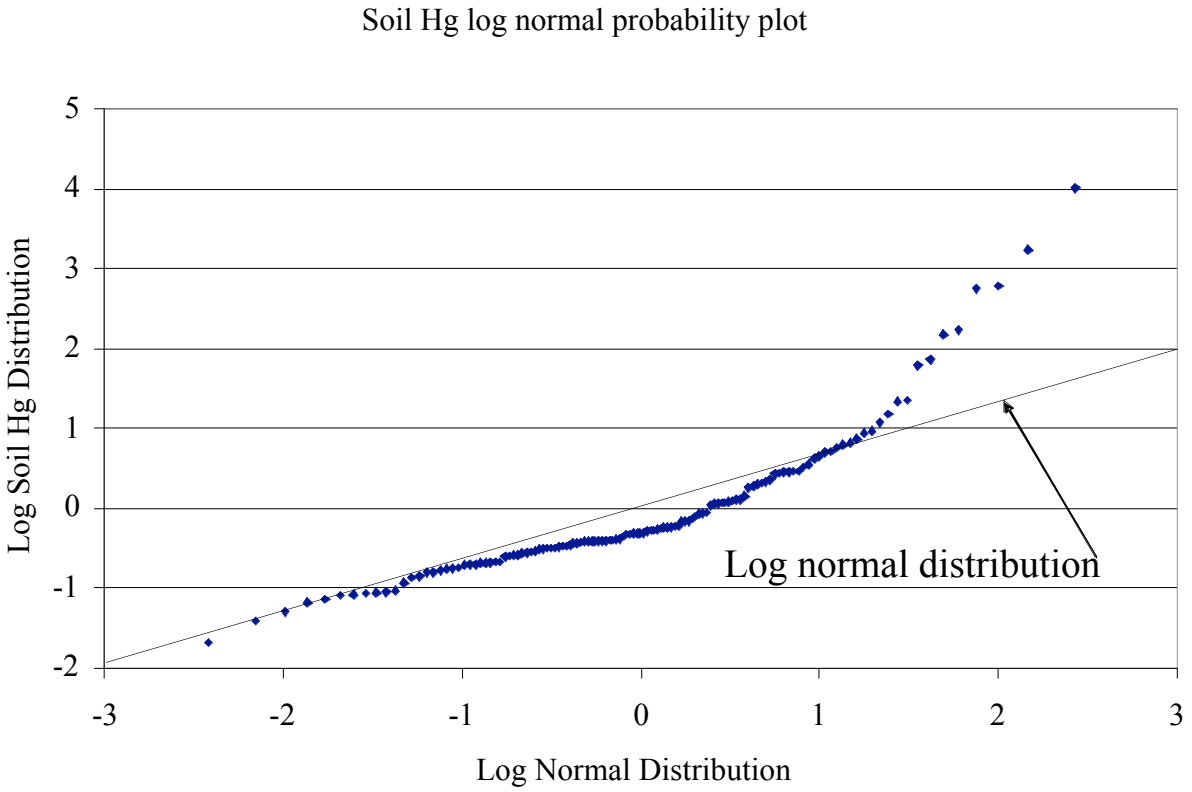

Figure 25 Log of all soil Hg concentration around the Kincaid plant versus log normal distribution.

In examining the data, it appeared that there were several outliers with values much higher than expected. For the purposes, of further analysis and data interpretation an outlier was defined as any location that had a soil mercury value that was more than a factor of 2 greater than at adjacent locations (< 1 mile). A factor of 2 difference from nearest neighbors could not occur due to deposition only because the deposition rate does not change by a factor of two in less than a mile, Figure 22. Therefore, the change is likely due to having different soil properties. This approach identified outliers at locations identified as A10, B2, D12, F2, and I10 on Figure 23. These sites had soil Hg values ranging from 56 – 155.6 ng/g. With the reduced data set, the average Hg value in the remaining 119 samples is 29.1 ng/g, a standard deviation of 8 ng/g and a range of 16.9 – 65.2 ng/g. The probability plot for the reduced data set, Figure 26, is markedly different than before with the highest measured values below the lognormal distribution curve.

There is a better match between the two distributions in Figure 26 than in Figure 25. However, it is not a particularly good match at the higher concentrations in either case.

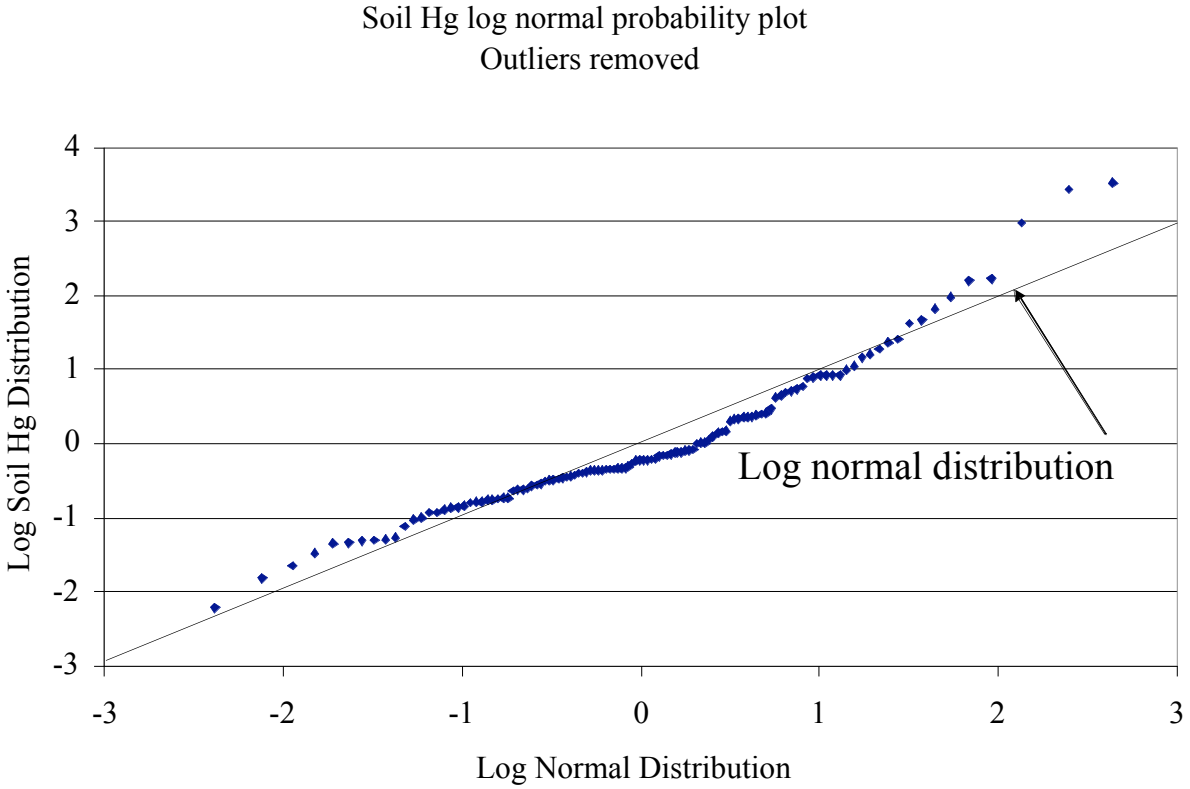

Figure 26 Log of soil Hg concentration without background locations and outliers versus log normal distribution.

Further analysis of potential "hot spots" was undertaken by focusing on the top 3 values, in relation to their immediate neighbors. These 3 values ranged from 2.6 to 5 times the median background (without its outlier). After averaging with up to 8 neighboring samples, these excess deposition ratios were reduced to 0.8, 0.9, and 1.5. Thus, only one location showed excess deposition, when averaged over a larger area to simulate effects on watersheds.

Comparison between the predicted deposition versus measured mercury concentrations in the soil was accomplished by overlaying the deposition map over the sampling map with sample results color-coded by measured concentration. Figure 27 is the graphical representation of the analysis for the samples near the Kincaid plant. Samples collected more than 10 miles from the plant for purposes of determining background are discussed later. Soil concentrations were binned into four approximately equal size groups containing 29 or 30 samples. Sample locations with symbols representing measured mercury levels represent the measured data. The range of the groups was 16.9 – 24.8, 24.8 – 27.1, 27.1 – 33.8, and 33.8 – 65.2 ng/g. The tight grouping of the middle groups around the average shows that 1/2 of the soil Hg measurements were in the range of 24.8 – 33.8 ng/g. Predicted regions of enhanced deposition are covered by the filled

contours with red representing 10 ug/m^2/yr, purple representing 5 ug/m^2/yr and blue representing 1 ug/m^2/yr.

Examining Figure 27 shows that there is not a particularly good correlation between regions of predicted enhanced deposition and soil Hg concentrations. Although higher concentrations do occur near the plant. In the region of predicted deposition in excess of 5 ug/m^2/yr, 6 of 12 sample locations have soil Hg concentrations in the top quartile. The highest measured concentration (with the outliers removed) 65.2 ng/g occurs at the location closest to the power plant (G7) and approximately 0.8 miles west of the plant.

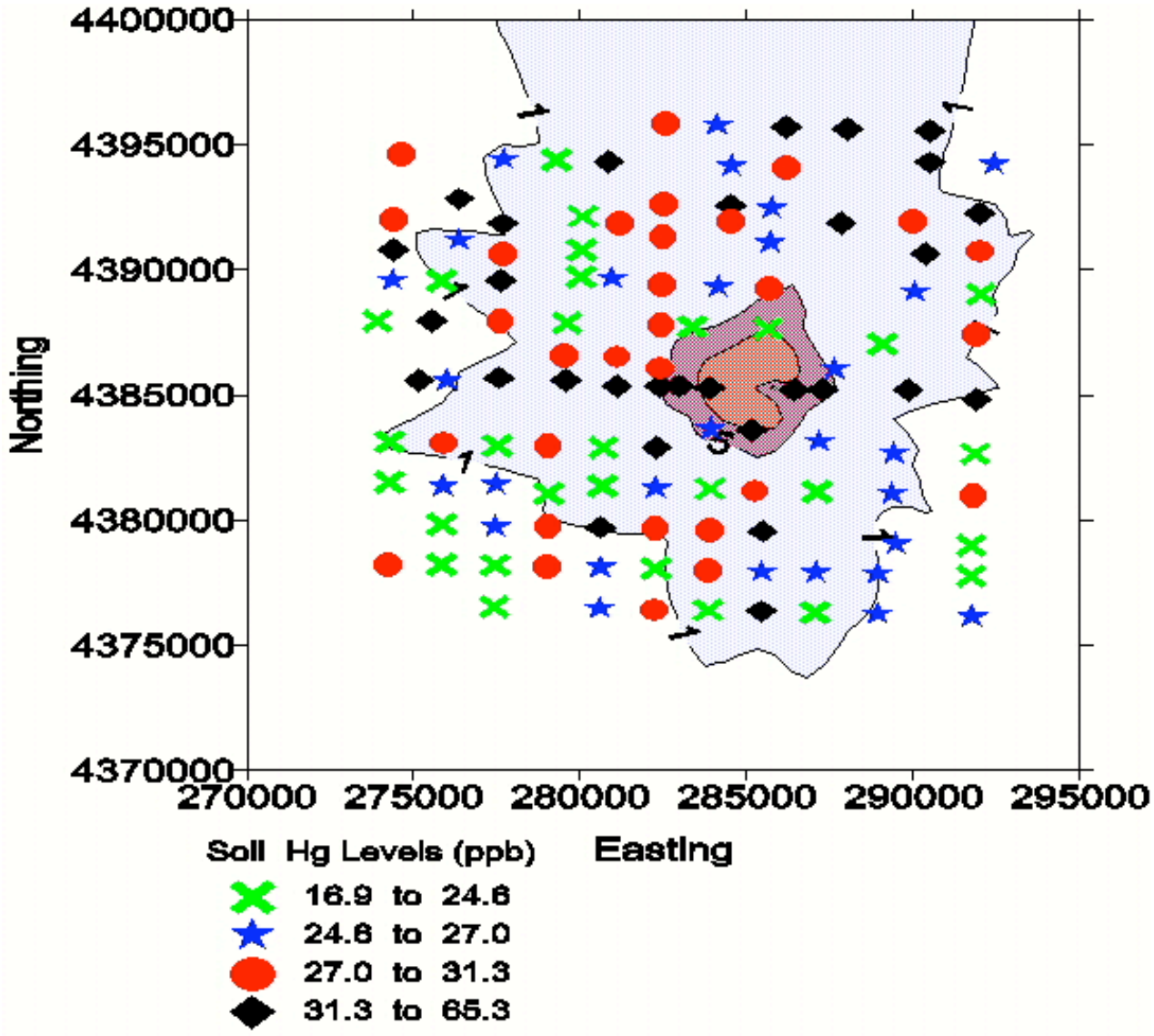

Figure 27 Comparison of measured soil Hg data and predicted Hg deposition (filled contours).

Figure 28 graphically represents the soil data, binned into the same four groups as in Figure 27 with a base map of the local roads. Examining this figure it appears that there is a correlation

between soil Hg and the East-West sample locations designated with the numeral 7 (e.g. A7, B7, etc.). This sampling transect was along the road that passes immediately south of the power plant. This road was the busiest road in the sampling domain near the plant as it had all of the employee traffic, it connected the two nearest towns, and had an access ramp to the interstate to the west of the plant. Also, samples in the lowest quartile were frequently associated with roads carrying less traffic.

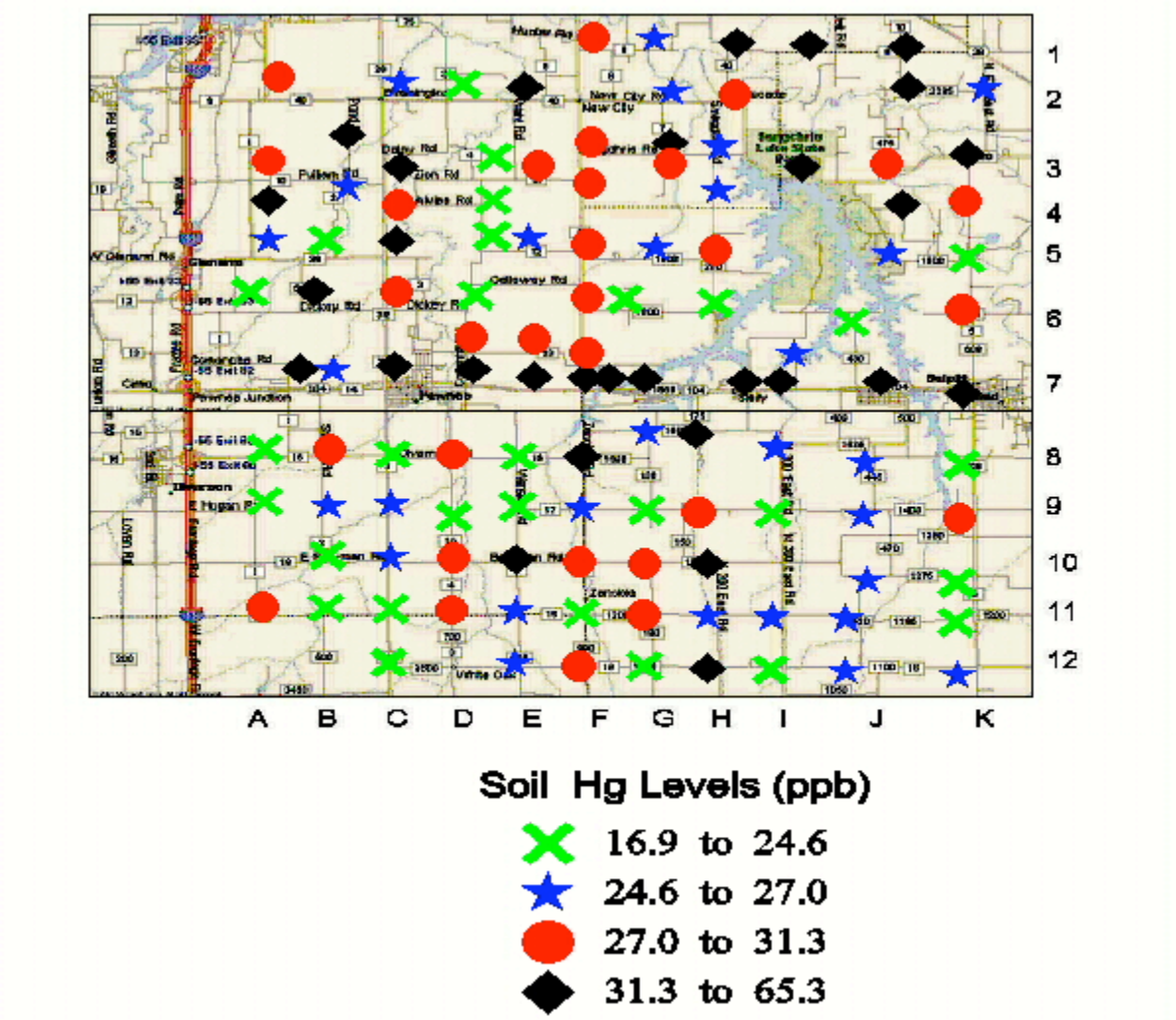

Figure 28 Soil Hg levels (ng/g dry) posted on local area map. Plant located between G7 and H7.

Table 5 presents the average mercury content (ng/g dry) of the three soil surface samples at each location. The outliers at A10 (56 ng/g) , B2 (155.7 ng/g), D12 (78.1 ng/g), F2 (78.9 ng/g), and I10 (120.5 ng/g) are not included. The average along each transect (A- K and 1 – 12) is also presented in the table. The plant was located between G7 and H7. High concentrations (65 ng/g at G7 and 42 ng/g at H7) were measured near the plant. The highest average transect is the east-west transect labeled 7 with an average value of 41 ng/g. The average for all soils is 29.1 ng/g.

There is no clear spatial pattern with distance from the plant in the transects. However, average concentrations were higher north of the plant (transects 1 -7), as compared to south of the plant.

Table 5 Average soil concentration (ng/g) dry at each sample location. Average values for the transects are the bottom and last rows of the table.

ID	A	B	C	D	E	F	G	H	I	J	K	Avg.
1						28.5	26.2	33.4	45.3	48.1		36.3
2	27.3		26.4	21.0	36.4		26.2	27.6		38.4	26.0	28.7
3	28.8	35.3	57.4	20.7	30.7	28.5	35.1	26.2	35.0	31.0	36.0	33.2
4	33.1	25.4	31.1	23.2		29.1	27.7	26.1		35.4	27.9	28.8
5	25.5	22.8	37.4	19.3	24.9	27.0	24.9	27.5		25.6	18.5	25.3
6	23.1	33.6	30.9	23.6		29.5	23.0	22.5		23.6	27.0	26.3
7A				27.0	30.6	27.0	32.9		26.9			28.9
7	34.1	24.6	39.2	35.3	63.9	33.8	65.2	42.1	35.4	37.8	39.7	41.0
8	23.9	30.9	23.3	27.4	23.6	43.7	26.3	47.8	25.7	25.0	23.9	29.2
9	23.7	26.1	25.9	20.9	20.0	26.7	20.7	27.4	24.5	25.3	28.5	24.5
10		20.8	24.6	30.8	31.3	27.8	29.6	31.3		26.3	21.1	27.1
11	27.9	22.3	21.8	27.1	26.1	24.5	29.4	25.6	25.9	26.2	22.8	25.4
12			16.9		25.2	27.7	23.8	31.8	23.9	26.1	25.3	25.1
Avg.	27.5	26.9	30.5	25.1	31.3	29.5	30.1	30.8	30.3	30.7	27.0	

Comparison of the surface (0- 5 cm) samples with the deep samples (5 – 10 cm) at the same locations suggests a strong correlation between the two, Figure 29. The highest variability occurred at high soil concentrations. However, the general trend was the same. The Spearman rank correlation coefficient between the surface and deep soils was 0.78 indicating a strong correlation. For these soils, statistical properties are in Table 6. The average of the deep soils was 0.6 ppb less than for the surface soils. However, this was not statistically significant.

Table 6 Comparison between surface and subsurface soil Hg concentrations .

	Surface (0 – 5 cm) (ng/g dry)	Subsurface (5 – 10 cm) (ng/g dry)
Minimum	12.0	15.3
Maximum	218.7	222.6
Average	32.0	31.4
Median	25.9	25.7
Standard Deviation	22.3	22.1

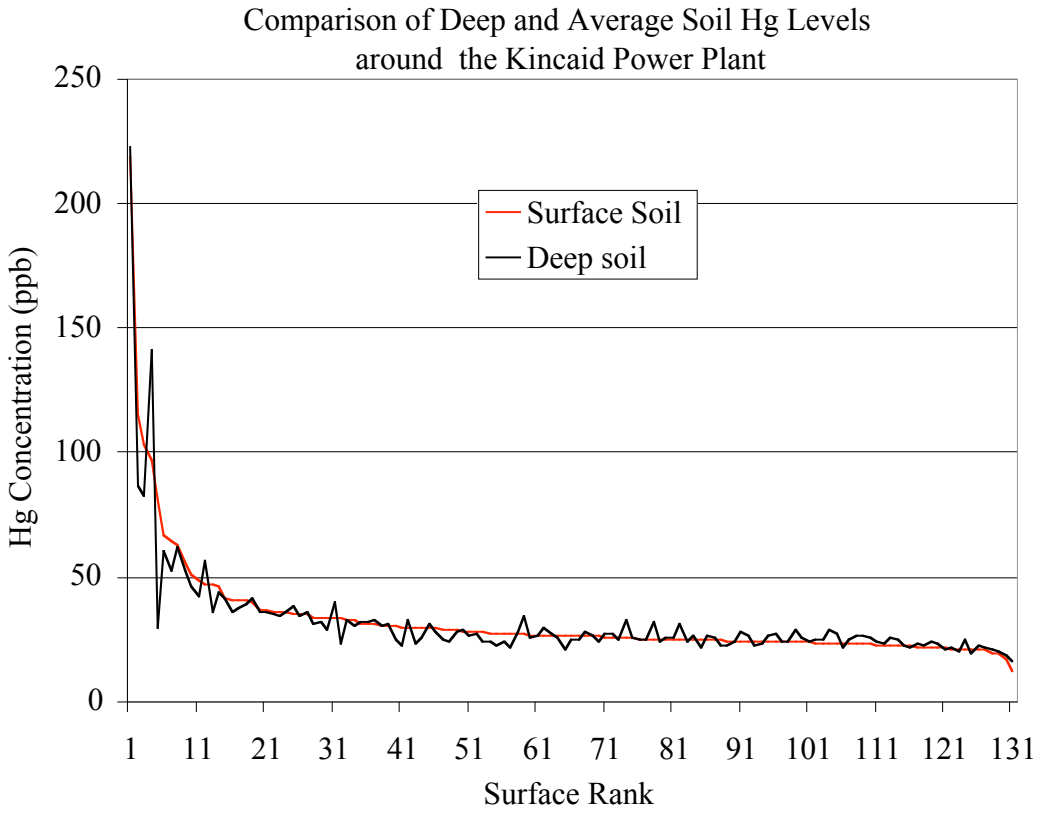

Figure 29 Comparison of surface and deep soil Hg levels around the Kincaid Power Plant.

3.4.4.2 Soil Background Samples

Attempts to define background were based on taking soil samples from eight locations at distances of 11 – 23 miles from the plant. Each location was predicted to be well outside the domain of influence for enhanced mercury deposition from the plant. The background values were generally higher than near the plant. Table 7 shows the measured values and distance from the plant. Table 7 also includes the nearest sampling location to the background site and the soil Hg level at this site, and distance from the road edge. The background sites had higher mercury levels than those near the plant with 4 of the 8 sites above 40 ng/g level. The background sites were selected on fairly well traveled roads for ease of access. Near the plant, higher mercury levels did seem to be associated with higher levels of traffic. The average of all background sites was 40.5 ng/g, close to the average value along the higher traffic transect 7, 41.0 ng/g. The locations may partially explain the higher mercury values associated with the background sites. The highest measured background site was also closest to the road being within 10 feet of the road. It should be noted that 10 samples near the plant were sampled within 10 feet of the road, however, exceptionally high values at these sites were not noted. In any event, the background samples could not be used to estimate a regional background.

Table 7 Background soil Hg levels (ng/g)

Location	Soil Hg (ng/g dry)	Nearest sampling location and soil Hg level (ng/g dry)	Distance from road edge (ft)	Distance to the plant (miles)
BG1	40.7	A2 (27.3)	25	17.4
BG2	24.8	G1 (26.2)	30	11.1
BG3	41.7	J1 (48.1)	15	17.9
BG4	92.3	K7 (39.7)	10	14.3
BG5*	20.3	K12 (25.3)	20	21.3
BG6	40.0	G12 (23.8)	15	15.8
BG7	38.6	A11(27.9)	20	23.1
BG8	25.4	A7 (34.1)	20	13.8

*BG5 had five samples between 19.5 and 20.6 ng/g and one sample at 168 ng/g. The one high value was treated as an outlier and not included in the averaging.

3.4.4.3 Vegetation Concentrations

One vegetation sample was collected at each sampling location. The vegetation samples are a measure of mercury deposition over the current growing season because the sample is from growth that occurred this year. Vegetation mercury levels are known to be influenced by both wet and dry deposition of mercury. Attempts were made to take the same type of vegetation from each location. This was not always possible, but all samples are grasses. If possible, samples from undisturbed vegetation were collected, however, in some cases, the vegetation near the edge of the road had been mowed. The samples were analyzed in duplicate and the average value was taken as a measure of the Hg content. For most samples, there was consistency between the two measurements. In contrast to Plant A, the vegetation samples did not show extremely high values and fit a log normal distribution well, Figure 30. Log normal distributions frequently provide a good representation of environmental data that is not impacted by outside sources. The statistics from the average at each location are in Table 8. The GSD is 2.0.

Table 8 Mercury concentration statistics for vegetation samples at Kincaid

	Value (ng/g)
Min	0.6
Max	22.5
Median	4.9
Average	5.6
Standard Deviation	3.8

The vegetation samples had a different spatial distribution than the soil samples. Figure 31 presents the measured mercury concentration in vegetation and the predicted deposition. Again, there is poor agreement between the predicted deposition and the measured vegetation mercury levels. Soil vegetation samples were divided into four groups representing the range from 0.6 – 3.1, 3.1 – 4.8, 4.8 – 7.0, and 7.0 – 22.5 ng/g. Each range had approximately 30 members and therefore, the ranges represent _ of all samples. Examining Figure 31 there does appear to be higher deposition values along the north-south axis centered on the plant, which is the direction of the prevailing winds, Figure 19. However, the highest values are not near the plant. In fact,

the samples with the two highest values were at locations B-8 (22.5 ng/g) and A-6 (22.3 ng/g). These locations are to the west more than 6 miles from the plant and not in the direction of the

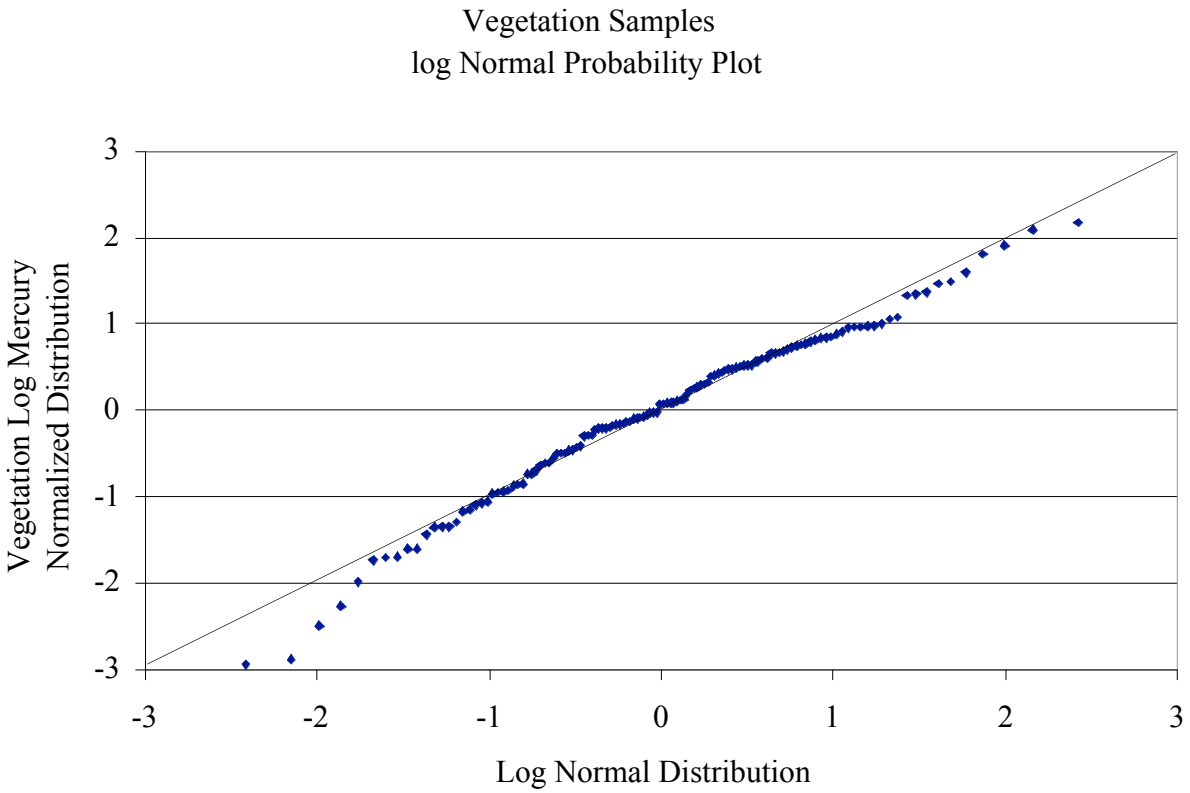

Figure 30 Probability plot for vegetation samples around the Kincaid Power Plant.

prevailing winds. Transect H, which samples directly north and south of the plant was characterized as having higher vegetation levels of mercury as compared to the other transects. Table 9 summarizes the average concentration data and provides the average along each transect. The average Hg vegetation value along transect H was 9.1 ng/g. No other transect had an average above 7.0 ng/g and the average of all samples was 5.6 ng/g. This could be interpreted as enhanced deposition during non-precipitation events. The rate of dry deposition was predicted to peak due north of the plant. Other major differences between the spatial distribution of Hg in soils and vegetation is the higher values noted along transect 7 in the soil were not found in the vegetation, and the soil concentrations tended to be higher to the northeast of the plant, while the vegetation samples had a higher average value southwest of the plant.

3.4.4.4 Vegetation Background Samples

Again, attempts to find a regional background for the site were unsuccessful. The eight background samples had a much higher average (7.0 ng/g) than the sites near the plant (5.6 ng/g). Five of the eight background samples had an average value greater than 8 ng/g. All of these samples are in the upper quarter of the distribution of samples near the plant. However, this is consistent with finding higher Hg levels in many of the background soil samples.

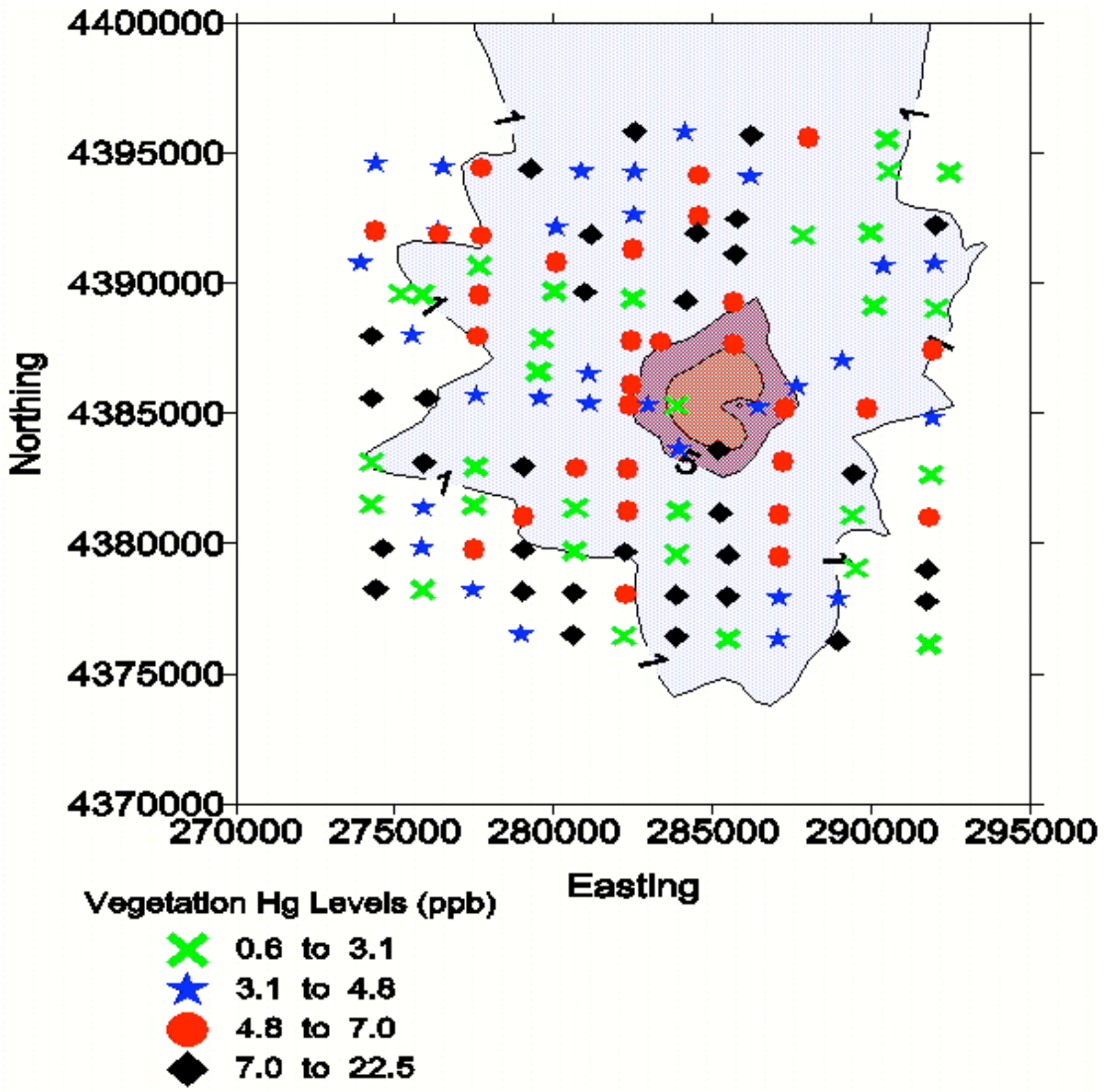

Figure 31 Vegetation Hg levels around the Kincaid Power Plant.

Table 9 Average Hg concentration in vegetation near the Kincaid Power Plant.

Location	A	B	C	D	E	F	G	H	I	J	K	Avg.
1						7.0	3.4	14.1	5.8	0.6		6.2
2	3.7	4.2	6.7	8.6	4.0	4.7	5.5	4.3		0.6	1.4	4.4
3	6.0	4.7	5.5	4.1	9.9	3.3	6.4	8.1	1.5	1.3	8.1	5.4
4	3.1	5.0	2.2	5.0		5.7	8.2	19.6		3.5	3.3	6.2
5	2.2	2.4	5.1	2.5	10.5	2.9	7.0	5.8		2.0	2.0	4.2
6	22.3	3.9	6.6	2.7		6.9	4.8	5.4		3.4	6.3	6.9
7A				1.0	5.1	5.5	3.2		4.7			3.9
7	8.4	7.7	3.8	3.8	4.7	6.1	1.7	4.2	6.5	5.6	4.5	5.2
8	0.9	22.5	1.2	10.6	3.9	6.1	3.6	11.3	5.0	7.6	1.5	6.7
9	2.4	3.8	1.8	5.9	2.3	6.8	2.4	14.8	6.5	2.4	5.9	5.0
10	13.0	4.7	5.0	8.0	2.0	7.2	7.4	8.8	5.2	1.9	7.9	6.5
11	8.3	2.9	4.4	9.8	7.0	6.6	11.3	10.7	4.2	3.7	7.9	7.0
12				4.4	9.2	1.9	7.6	2.6	4.6	10.7	2.9	5.5
Average	7.0	6.2	4.2	5.5	5.8	5.4	5.6	9.1	4.9	3.6	4.7	

3.4.5 Comparison of Mercury Concentrations as a function of Distance

The data were grouped as a function of distance from the plant in one mile intervals, Table 10. All vegetation samples were used in the analysis. The sites determined to be outliers in the previous section were not used in the soil analysis. There is a correlation with distance for the soil samples, with higher values measured closer to the plant. There is no apparent correlation with distance for the vegetation samples. If locations with vegetation mercury levels that are more than twice the value of any nearest neighbor are treated as outliers, four samples would be omitted (A6, B8, H1, and J12 in Table 9). These samples are located between five and eight miles from the plant and they change the profile such that the vegetation profile peaks between two and four miles and then decrease monotonically until the last interval at 8 – 9 miles, which remains unchanged. This interval has only 3 data points and the results are less reliable than in the other section with more data.

Table 10 Soil and vegetation mercury concentrations as a function of distance.

Distance (miles)	# Samples	Average Soil Hg (ng/g)	# Samples	Average Vegetation Hg (ng/g)
0 – 1	2	38.7	2	5.3
1 – 2	7	35.3	9	5.2
2 – 3	15	30.4	13	6.1
3 – 4	12	27.1	14	6.1
4 - 5	25	27.4	26	5.4
5 - 6	19	28.3	23	5.2
6 - 7	22	30.4	22	5.8
7 - 8	12	26.9	12	6.2
8 - 9	3	29.3	3	6.0

3.4.6 Comparison of Soil Sampling Results at the Kincaid Power Plant.

In the April 1974 the Illinois Natural History Survey (Anderson, 1977) conducted a soil sampling campaign on a 3.2 km (2 mile) square grid, 38.6 km across with the Kincaid plant at the center. Two soil types Sable (Illiopolis) silty clay loam (north of the plant) and Virden silty clay loam (south of the plant) were observed. Samples were taken from the top two cm in agricultural fields that had not been disturbed for several months. These sites were at least 50 m and usually 100 m from the road. In the current study, a 1.6 km grid, 20 km wide centered around the plant was used. Soil samples were collected in undisturbed regions generally about 20 feet from the road. However, in some cases, this was not possible and 10 samples were collected at 10 feet or less from the road edge. Samples were homogenized over the top 5 cm. Moisture content was measured for all samples and mercury content was reported on a dry weight basis. The previous study reported mercury concentrations on a wet weight basis. The average correction factor for converting from wet weight to dry weight basis was 1.25. Therefore, in the following comparison, all values measured in the 1974 study were multiplied by this factor to have a uniform basis for comparison.

In the 1974 study, the Hg concentration values ranged from 1 – 50 ng/g dry weight basis. This is similar to the current study (16.9– 65 ng/g) after discounting the outliers. High values as found in the outliers (up to 155 ng/g) were not observed in the previous study. The 1974 study did not

publish all of the data however, they provided average values for four quadrants (NE, NW, SE, and SW). Table 11 presents the average values in these regions from the 1974 and current studies. Both studies show the same relative ranking of quadrants with the NE being the highest followed by NW, SW and SE. The prevailing winds are from the south to the north so this may reflect enhanced deposition. However, the 1974 study reported that there was a slight change in soil type between north and south of the plant, which also could be responsible for the difference. The values for the 2004 study were approximately 25% higher than the 1974 study. This could be due to many different factors other than deposition including analytical techniques, sampling locations, and sampling protocol. The 1974 study was taken from the agricultural fields that are plowed to a depth of 17 cm annually. This would cause mixing and presumably dilution as deposition is a surface process. Mercury content typically decreases with depth from the surface. Also, in the current study an apparent correlation with vehicular traffic along the road was observed and perhaps the difference in average values reflects the difference in sample location (roadside versus field). In any event, it is not possible to determine if the difference between average Hg values in the two studies is meaningful.

Table 11 Average soil mercury concentrations by quadrant.

Quadrant	Average Soil Hg (ng/g dry) 2004 Study	Average Soil Hg (ng/g dry) 1974 Study	Difference from SW Quadrant (ng/g)	
			1974	2004
SW	25.8	18.8	----	----
SE	26.8	20.0	1.2	1.0
NW	30.9	23.8	5.0	5.1
NE	31.8	27.5	8.7	6.0

3.4.7 Estimated Mass Deposition

Mass deposition estimates were performed for the approximately 400 square kilometer sampling region around the plant. Using a soil density of 1.5 g/cm^3 and limiting the excess deposition to the top 5 cm gives a total volume of soil of 2.0 10^7 m^3 and a soil mass of 3.0$10^{13}$ g over the study area. Table 12 presents the excess estimated amount deposited for four cases:

a) using the soil surface average minus the soil subsurface average (0.6 ng/g),
b) the soil surface average minus the value of the soil concentration where 2/3 of the measured values were greater than the measured value (3.7 ng/g),
c) soil average by quadrant minus the lowest soil average by quadrant (Table 11), this approach was used in the 1974 study by Anderson (Anderson, 1977), this approach assumes that the SW quadrant (lowest average quadrant) is not impacted by plant emissions and is representative of background.
d) the estimate based on deposition modeling using ISCST (790 g/yr) multiplied by the operational period (37 years).

Table 12 contains the estimate of deposited mass, the % of RGM emissions represented by this mass, and the % of total emissions that this represents. Total emissions were assumed to be 160 kg/yr multiplied by the 37 year operational period of the plant. Twenty percent of emissions were assumed to be RGM. Total emissions are likely to be higher as an emission estimate of 530 kg was suggested during the early 1970's (Anderson, 1977). Also, prior to 1994 when locally mined coal was burned, the percentage of RGM was likely to be higher than 20%. Both of these assumptions skew the estimate of the % of mercury deposited to higher values. The table shows

that for any scenario, less than 9 % of the RGM and less than 2% of the total mercury emissions from the plant deposited within a 20 km square centered on the plant. This analysis also supports the contention that although mercury levels may be slightly elevated near the plant, a 'hot spot' with substantially elevated levels does not occur. The analysis also suggests that greater than 90% of the mercury emitted from the plant enters the regional or global mercury cycle.

Table 12 Mercury mass deposition estimates around the Kincaid Plant based on soil data and ISCST modeling

	Excess (ng/g)	Total Mass Deposited (kg)	% of RGM	% of Hg(total)
Case A: Surface avg. minus subsurface avg.	0.6	16.4	1.4	0.28
Case B: Surface average minus soil concentration at 1/3 of the distribution	3.7	101	8.5	1.7
Case C: Quadrant average minus lowest quadrant average	1.0 (SE-SW) 5.1 (NW-SW) 6.0 (NE-SW)	6.8 34.8 41.0 Total – 82.6	7.0	1.4
ISCST Deposition	N/A	29.2	2.5	0.5

The previous study (Anderson, 1977) estimated that "more than 26% and possibly as high as 70% of the mercury emitted by the Kincaid plant" was deposited within a 19.3 km radius of the plant. The cause for this discrepancy in this estimate and the estimates in Table 12 lies in the differing assumptions made in the two analyses. In the 1977 study, the volume of soil was estimated using a depth of 17 cm, the plow depth, even though the samples were only from the top 2 cm. In addition, in that study, the background value was set to the lowest value in the four sampled quadrants. In any event, the 1977 estimates appear to be biased high. If the current emission rate of 160 kg/yr is used to estimate the lifetime of the plant emissions, deposition of 26% of the Hg within 19.3 km radius would lead to an increase in average soil concentration in the top 5 cm of 17.6 ng/g. If this occurred, the average soil Hg concentrations on a dry weight basis would be expected to be around 35 – 40 ng/g, which is substantially higher than observed. In addition, Hg emissions rates in the early part of the operating history of the plant tended to be as much as a factor of three higher than the value used to estimate the amount of Hg that could be deposited. If the total emissions were higher than estimated, the increase in soil concentration would be expected to be even higher.

Assuming that plant emissions averaged 160 kg/yr over the 37 year operational period, 5920 kg of mercury would have been emitted by the plant. If all of this mercury deposited within the 19.3 km radius of the plant and was retained in the top 5 cm of soil, the average soil Hg content would increase by 67 ng/g. This has not happened, which further suggests that deposition in this region is less than 10% of the total emissions.

4.0 Framework for Analyzing the Local Impacts of Mercury Emissions on Public Health

The modeling and data both suggest that emissions from a single coal-fired power plant will increase (>10%) deposition over a region less than 25 km from the plant. Rigorous assessment of the impacts of potential cap-and-trade scenarios for mercury emissions from coal-fired power plants will require modeling as it is not feasible to collect data from around each plant. A potential framework for understanding the impacts of local mercury deposition is outlined below. The steps include:

- Define mercury emissions.
  - Identify power plants and relevant factors for mercury deposition (type of coal, stack data (height, exit velocity, exit temperature, exhaust area), mercury emission rates, mercury speciation in emissions.
- Estimate mercury deposition.
  - Obtain meteorological data (hourly data on wind speeds and directions, precipitation rates, etc.)
  - Model deposition based on power plant and meteorological data. Focus on increased deposition near the plant.(1-25 km).
- Estimate potential for increased exposure through the fish pathway.
  - Identify water bodies within 25 km of the plant.
  - Obtain fish mercury levels in fish in nearby water bodies from EPA and state data
  - Obtain information on local fish consumption patterns.
  - Obtain population data by county
  - Using fish mercury, fish consumption and population data estimate exposure.
- Calculate increased risks
  - Use Dose-response curves to translate exposure to risk.
  - Calculate risks for general population and susceptible groups (consumers of local fish)

This would be a huge labor intensive task for the more than 1000 coal-fired power plants in the U.S. Prioritization could be performed based on the size of the plant, local population, and local fish consumption patterns to estimate the plants with the highest risk of causing an effect.

4.1 Define Mercury Emissions

A number of data bases exist that contain information about power plants and their emissions. The DOE data base on Steam-Electric Plant Operation and Design Data, EIA-767, (http://www.eia.doe.gov/cneaf/electricity/page/eia767.html) contains plant location, including stack height, area at top of stack, exit temperature at 100% power, exit velocity at 100% power, and primary fuel. The EPA's Toxic Release Inventory data base (www.epa.gov/tri/) contains total mercury emissions but does not include speciation data. This data base is difficult to use for our purposes as the analyst needs to search through and select out only coal-fired power plants from all of the various emission sources. Considerable effort was necessary to strip the huge amount of other industries out to obtain the power plant data. A more direct approach is to use the compilation of total mercury emission rates by power plant from http://www.epa.gov/ttn/atw/combust/utiltox/pltxplt2.pdf.

To confirm the location of the power plants web based utility such as TerraServer (http://www.terraserver-usa.com/) and MapQuest (www.mapquest.com) were used.
In some cases, lat-long data were available and these could be input directly into the above utilities. TerraServer provides aerial photos and USGS topo maps. The plant stacks could often, but not always, be seen in the aerial photos. Power plants are marked on the USGS topographic maps, so it was possible to know one had found a power plant. The most difficult cases to identify were the newer plants, which were not on the map. In some cases these were identified through other sources, such as web pages of the facility. In some cases, the directions were not specific, for example "9 miles north of Jewett." TerraServer® 6.0 was helpful in the location of these plants effort since it can flip back and forth from topographic maps to aerial photos.

In 1999 EPA issued an Information Collection Request on mercury speciation from coal-fired power plants. Two hundred forty mercury speciation measurements were performed at more than 60 plants and the results are summarized at:
http://www.epa.gov/ttn/tw/combust/utiltox/rawdata1.xls. This information was also used as the basis for estimating mercury speciation at coal plants where measurements are not readily available.

4.2 Estimate Mercury Deposition

With the emissions data and plant factors, deposition can be modeled using local meteorological data. Lat-Long data for the plants can be used to determine the nearest airport that has the necessary data for modeling deposition. There are a number of sources for this information. There are also a number of formats for storing meteorological data. The choice of software used in the deposition analysis affects the selection of the source for meteorological data. For the analysis performed in Section 3 using ISCST, data were downloaded from WebMET http://www.webmet.com/. Other sources of data include EPA, http://www.epa.gov/scram001/tt24.htm and NOAA has data for a nominal charge, http://www.ncdc.noaa.gov/oa/wmo/wdcamet.html. The data include hourly information on wind speed and direction, precipitation rates and type (rain, freezing rain, snow, etc.), and other parameters such as ceiling height, dew point temperature, direct solar radiation, global solar radiation, station pressure, relative humidity, sea level pressure, dry bulb temperature, wet bulb temperature, leaf area index, and total sky and total opaque sky cover.

The data in section 3 and found in the literature, Section 2, suggest that substantially increased mercury deposition leading to elevated fish Hg concentrations will not occur. Modeling and data suggest about a 10 – 20% increase in deposition within a few km of the plant and lower values at further distances.

4.3 Estimate Potential for Increased Exposure

Fish consumption is the major pathway for mercury to enter the body. Therefore, human exposure depends on the number of people eating fish, the amount of fish they consume, and the levels of mercury in the fish. The major health risk is to the developing fetus as mercury is a neurotoxin that can cross into the developing fetal brain. This leads to a focus on women of child bearing age when assessing human health risks.

For a given power plant, the number of people eating locally caught fish will depend on the local population and the availability of fishing locations. The local population can be estimated from census data. The 1999 census data for Beaver County, PA where the Bruce Mansfield power plant is located had 183, 417 persons. There were 97,432 females of which 80,501 were between the ages of 18 – 49 (roughly the span of child-bearing years). The Bruce Mansfield plant is located on the Ohio River and 62 water bodies are located within 25 km of the plant. Most of these are small streams that will not support large quantities of fish. Appendix A lists the water bodies around the Bruce Mansfield plant.

Ideally, information would be available on the mercury levels in fish around the power plant. Most states, including Pennsylvania, have active fish sampling programs. The Pennsylvania data base was reviewed and information on four species (channel catfish, common carp, small mouth bass and large mouth bass) n the 62 water bodies within the 25 km of the plant were found. Mercury levels were not unusually high with the highest value of 0.42 mg/kg for large mouth Bass (Appendix A). There were no advisories for fish consumption in Beaver County for mercury although there was one water body with an advisory for PCB contamination.

Use of data from four water bodies and four species of fish to represent the mercury levels in the area may not be representative. In this case, the states data base or the EPA data base (EPA, 1999) could be used to estimate mercury concentrations in fish. Increased mercury deposition from the plant would presumably lead to increased mercury levels in fish. To adjust for this, some assume that the increase in fish mercury level is linearly proportional to the amount of deposition [Sullivan, 2003]. However, the review of the literature in section 2 suggests that a square root dependence better fits the data. The transformation of deposited mercury to methyl mercury and its incorporation into the food chain is complex depending on water chemistry, microbial reactions, and uptake. Thus, it is currently not possible to determine the response of any water body to increased deposition with a high degree of confidence. However, most agree that a linear increase in fish mercury concentration with mercury deposition should provide an upper bound. Thus, the excess deposition could be used to scale the mercury levels in fish.

The last component needed to estimate exposure is the consumption rate of locally caught fish. National surveys of fish consumption suggest that women aged 15 to 44 years, the childbearing years, consume a mean daily average of 4.3 grams of "as prepared" freshwater and estuarine finfish and shellfish. This same group consumed, on average, 7.0 ± 0.9 grams/person/day of "as prepared" marine finfish and shellfish. Again, it would be ideal if data were available around the power plant and for people that consume large amounts of fish. This is generally not the case. However, there have been a number of studies on fish consumption in selected regions. A detailed study was performed for fishers from the Santa Monica pier with median consumption around 25 g/d and 90% of the fishers consuming less than 100 g/d. The fraction of the total population that is represented by this subset is not known, but it is clearly small (< 1%) or the national averages would be higher. In any event, there are small groups of people that will consume large amounts of fish. The issue for assessing local impacts of mercury deposition from coal-fired power plants becomes one of identifying these groups and determining how much of their diet comes from locally caught fish. For most power plants, this information will be unknown and could only be obtained through labor intensive surveys. Therefore, values found in the literature may be the best available approach for estimating local impacts. Attempts to match the power plant to data collected from the region will lead to more credible analysis. For example, data from New Jersey is likely to be more relevant than data from California when looking at consumption patterns for locally caught fish. Similarly, data from populations fishing

in rivers and streams is more appropriate for the Bruce Mansfield region as compared to data from coastal regions with saltwater.

4.4 Calculate Risks Based on Increased Exposure

The population risk of a health effect is estimated as the sum of the products of the incremental probability of exposure at a given level for each member of the population times the probability of experiencing the effect at that exposure level. The health effect is damage to the developing fetal brain, which is measured in children using standard neurological tests (e.g., memory skills, coordination, dexterity, etc.). In previous studies (Sullivan, 2002; Sullivan, 2003) a Monte Carlo approach is used that samples among the distribution of consumption behavior and the distribution of mercury concentrations in fish. The result is a distribution of daily intake (i.e., 3% of the population has an intake of 0.1 µg/d, 5% has an intake of 0.2 µg/d, and so on). Dose response factors can be used to estimate the risk based on consumption (Sullivan, 2002). The previous studies were designed to examine risk on a regional basis and concluded that the increase in risk due to the mercury emissions was small. For deposition within a few km of the power plant, deposition rates could be higher than for the estimates based on a regional scale.

5. Conclusions

A thorough quantitative understanding of the processes of mercury emissions, deposition, and translocation through the food chain is currently not available. Complex atmospheric chemistry and dispersion models are required to predict concentration and deposition contributions, and aquatic process models are required to predict effects on fish. However, there are uncertainties in all of these predictions. Therefore, the most reliable method of understanding impacts of coal-fired power plants on Hg deposition is from empirical data.

A review of the literature on mercury deposition around sources including coal-fired power plants found studies covering local mercury concentrations in soil, vegetation, and animals (fish and cows). There is strong evidence of enhanced local deposition within 3 km of the chlor-alkali plants, with elevated soil concentrations and estimated deposition rates of 10 times background. For coal-fired power plants, the data show that atmospheric deposition of Hg may be slightly enhanced. On the scale of a few km, modeling suggests that wet deposition may be increased by a factor of two or three over background. The measured data suggest lower increases of 15% or less. The effects of coal-fired plants seem to be less than 10% of total deposition on a national scale, based on emissions and global modeling.

The following summarizes our findings from published reports on the impacts of local deposition. In terms of excesses over background the following increments have been observed within a few km of the plant:

- local soil concentration Hg increments of 30%-60%,
- sediment increments of 18-30%, and
- wet deposition increments of 11-12%.
- fish Hg increments of about 5-6%, based on an empirical finding that fish concentrations are proportional to the square root of deposition.

Important uncertainties include possible reductions of RGM to Hg_0 in power plant plumes and the role of water chemistry in the relationship between Hg deposition and fish content.

Soil and vegetation sampling programs were performed around two mid-size coal fired power plants. The objectives were to determine if local mercury hot-spots exist, to determine if they could be attributed to deposition of coal-fired power plant emissions, and to determine if they correlated with model predictions. These programs found the following:

- At both sites, there was no correlation between modeled mercury deposition and either soil concentrations or vegetation concentrations. At the Kincaid plant, there was excess soil Hg along heavily traveled roads. The spatial pattern of soil mercury concentrations did not match the pattern of vegetation Hg concentrations at either plant.
- At both sites, the subsurface (5 – 10 cm) samples the Hg concentration correlated strongly with the surface samples (0-5 cm). Average subsurface sample concentrations were slightly less than the surface samples; however, the difference was not statistically significant.
- An unequivocal definition of background Hg was not possible at either site. Using various assumed background soil mercury concentrations, the percentage of mercury deposited within 10 km of the plant ranged between 1.4 and 8.5% of the RGM emissions. Based on computer modeling, Hg deposition was primarily RGM with much lower deposition from elemental mercury. Estimates of the

percentage of total Hg deposition ranged between 0.3 and 1.7%. These small percentages of deposition are consistent with the empirical findings of only minor perturbations in environmental levels, as opposed to "hot spots", near the plants.

The major objective of this study was to determine if there was evidence for "hot-spots" of mercury deposition around coal-fired power plants. Although the term has been used extensively, it has never been defined. From a public health perspective, such a "hot spot" must be large enough to insure that it did not occur by chance, and it must affect water bodies large enough to support a population of subsistence fishers. The results of this study support the hypothesis that neither of these conditions has been met.

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6. References

Abbott, M.L., D.D. Susong, M. Olson, and D.P. Krabbenhoft, Mercury in soil near a long-term air emission source in southeastern Idaho, Environmental Geology (2003) 43:352-356.

Anderson, W.L., Smith, K.E., Dynamics of Mercury at a Coal-Fired Power Plant and Adjacent Cooling Lake, Environ Sci Tech,(1977),11:75-80.

Biester, H, G. Muller, and H.F. Scholer, Estimating distribution and retention of mercury in three different soils contaminated by emissions from chlor-alkali plants, Science of Tot Env (2002a),284:177-189.

Biester, H, G. Muller, and H.F. Scholer, Binding and mobility of mercury in soils contaminated by emissions from chlor-alkali plants, Science of Tot Env(2002) 284: 191-203.

Cox, W. M., H. W. Rorex, and G. K. Moss, *Evaluation of Rural Air Quality Simulation models, Addendum C: Kincaid SO_2 Data Base*, EPA-450/83-003c, U.S. Environmental Protection Agency, Research Triangle Park, NC, March 1986.

Crockett, A., Kinnison, R.H., Mercury Residues in Soil around a Large Coal-Fired Power Plant, Environ Sci Tech (1979) 13:712-5.

Edgerton. E. B. Hartsell, and J. Jansen, "Hg Speciation in Coal-Fired Power Plant Plumes: Evidence for Reduction of RGM to HG(0)," 7th Annual Conference on Mercury as a Global Pollutant, Ljubljana, Slovenia, July 2, 2004.

U.S. Department of Energy, Energy Information Administration, "Steam-Electric Plant Operation and Design Data," EIA-767, (http://www.eia.doe.gov/cneaf/electricity/page/eia767.html).

U.S. Environmental Protection Agency, User's Guide for the Industrial Source Complex (ISC3) Dispersion Models, Volume I – User Instructions, EPA-454/B-95-003A, Office of Air Quality Planning and Standards Emissions, Monitoring, and Analysis Division, Research Triangle Park, NC. 1995

U.S. Environmental Protection Agency, Fate and Transport of Mercury in the Environment, Mercury studyReport to Congress, vol. III, EPA-452/R-970995, 1997.

U.S. Environmental Protection Agency, National Survey of Mercury Concentrations in Fish: Data Base Summary 1990 – 1995, EPA 823-R-99-014 1999.

U.S. Environmental Protection Agency, State Mercury Emissions Estimates from Power plants, June 2001, http://www.epa.gov/ttn/atw/combust/utiltox/utoxpg.html#DA1.

Environmental Science and Technology Science Page, "Controversial Results Downplay Power Plant Mercury Emissions," Nov. 3, 2004. http://pubs.acs.org/subscribe/journals/esthag-w/2004/nov/science/rr_mercury.html

Environmental Science and Technology Science Page, "Mercury Trading Scheme Raises Concern," March 4, 2004. http://pubs.acs.org/subscribe/journals/esthag-w/2004/mar/science/kc_mercury.html

Environmental Science and Technology Policy Page, "Mercury Policy Debate Heats Up," July 28, 2004. http://pubs.acs.org/subscribe/journals/esthag-w/2004/jul/policy/kc_mercury.html

Gustin, M.S., J.A. Ericksen, D.E. Schorran, D.W. Johnson, SE. Lindberg, and J.S. Coleman, Application of Controlled Microcosms for Understanding Mercury Air-Soil-Plant Exchange, Environ Sci Tech, (2004), 38:6044-6050.

Klein, D.H., Russell, P., Heavy Metals: Fallout around a Power Plant, Environ Sci Tech (1973) 7:357-8.

Kotnik, J., Horvat, M., Mandic, V., Logar, M., Influence of the Sostanj coal-fired thermal power plant on mercury and methyl mercury concentrations in Lake Velenje, Slovenia, Science Total Environ (2000), 259:85-95.

Landis, M.S., Vette, A.F. Keeler, G.J., Atmospheric Mercury in the Lake Michigan Basin: Influence of the Chicago/Gary Urban Area, Environ Sci Tech (2002a) 36: 4508-17.

Landis, M.S., Keeler, G.J., Atmospheric Mercury Deposition to Lake Michigan during the Lake Michigan Mass Balance Study, Environ Sci Tech(2002b), 36:4518-24.

Laudal, D., G. Dunham, L.Levin, Mercury Control Technology R&D Review, Pittsburgh, PA, July 14 – 15, 2004.
http://www.netl.doe.gov/publications/proceedings/04/HgReview/Laudal2.Presentation%20for%20Project%20Review%20Meeting%20Plume%20Study%20-%20Pittsburgh%20July%2014-15%202004%20-%202.pdf

Lodenius, M. Dry and wet deposition of mercury near a chlor-alkali plant, Science Tot Environ (1998), 213: 53-56.

Lopez Alonzo, M., Benedito, J.L., Miranda, M., Fernandez, J.L., Castillo, C., Hernandez, J., Shore, R.F., Environ Pollut (2003), 125:173-81.

Mercury Deposition Network, http://nadp.sws.uiuc.edu/mdn.

Department of Health and Human Services, Centers for Disease Control and Prevention, Second National Report on Human Exposure to Environmental Chemicals, 2003. http://www.cdc.gov/exposurereport/2nd/pdf/secondner.pdf

Menounou, N., Presley, B.J., Mercury and other trace elements in sediment cores from central Texas lakes, Arch Environ Contam Toxicol (2003), 45:11-29.

Nadim, F., Perkins, C., Liu, S., Carley, R.J., Hoag, G.E., Chemosphere (2001), 45:1033-43.

Pinkney, A.E., Logan, D.T., Wilson, H.T., Arch Environ Contam Toxicol (1997), 33:222-9.

Risch, M., in Proceedings. Great Lakes Regional Workshop, Measuring Atmospheric Mercury: Goals, Methods, and Results. March 26, 2003. Available at http://www.deq.state.mi.us/documents/deq-aqd-toxics-HgWorkshop.doc

Seigneur, C., Vijayaraghavan, K., Lohman, K., Karamchandani, P, Scott, C., Global Source Attribution for Mercury Deposition in the U.S., Environ Sci Tech (2004), 38:555-569.

Sensen, M., and D.H.S. Richardson, Mercury levels in lichens from different host trees around a chlor-alkali plant in New Brunswick, Canada. , Science Tot Environ (2002),293:31-45.

Southworth, G.R., Lindberg, S.E., Zhang, H., Anscombe, F.R., Fugitive mercury emissions from a chlor-alkali factory; sources and fluxes to the atmosphere, Atmospheric Environment (2004) 38:597-611.

Sullivan , T.M., F.W. Lipfert, S.C.. Morris, P.D. Moskowitz, Potential Health Risk Reduction Arising From Reduced Mercury Emissions from Coal-Fired Power Plants, FY 2001 Report, September, 2001, BNL-71538-2003.

Sullivan, T.M., Lipfert, F.D., Morris, S.M., The Local Impacts of Mercury Emissions from Coal Fired Power Plants on Human Health Risk, Progress Report for the Period of March 2002 – March 2003, BNL-71554-2003, May 2003.

Susong, D.D., M.L. Abbott,and D.P. Krabbenhoft, "Mercury accumulation in snow on the Idaho National Engineering and Environmental Laboratory and surrounding region, southeast Idaho, USA, Environmental Geology (2003), 43:357-363.

Tack, F.M.G., T. Vanhaesebroeck, M.G. Verloo, K. Van Rompaey, E. Van Ranst, Mercury baseline levels in Flemish soils (Belgium), " Environmental Pollution (2005),134:173-179.

Wang, D., Shi, X., Wei S., Accumulation and transformation of atmospheric mercury in soil, Science Tot Environ (2003), 300:209-214.

Xiao, Z., J. Smar, O. Lindqvist, E. Giouleka, Atmospheric mercury deposition to grass in southern Sweden, Science Tot Environ (1998),213: 85-94.

Appendix A: Data Relevant for Risk Assessment near the Bruce Mansfield Power Plant

Using the Bruce Mansfield plant, data were extracted from the Energy Information Administration Steam-Electric Plant Operation and Design Data, EIA-767, (http://www.eia.doe.gov/cneaf/electricity/page/eia767.html). These data are from 2000. Data extracted from this data base that are important to evaluating health impacts include:

- Fuel: Bituminous coal
- Lat 40^{o} 38' 03" Long 80^{o} 24' 59"
- Located in Beaver County, PA, 25 miles northwest of Pittsburgh. Zip 15077
- Source of water: Ohio River.
- No fly ash injection
- NOx control operative
- Low NOx process is low NOx burner
- Low NOx process is Overfire air
- NOx removal factor 0.39
- Mercury control (none)
- Stack Height: 600 ft
- Area at top of stack 284 sq ft.
- Exit rate 130,500 (at 100%) , 732,000 (at 50%)
- Exit temperature 126 (at 100%), 119 (50%)

A.1 Speciation of Mercury Emissions in Bruce Mansfield

As part of the 1999 EPA information collection request, speciation data were obtained for the Bruce Mansfield Plant. This data is reproduced in the table below. This data was used in modeling deposition around the plant (Sullivan, 2003).

Table A-1 Mercury Speciation from Bruce Mansfield.

	Hg particle g/y	Hg Reactive g/y	Hg Elemental g/y	Hg total kg/y
Bruce Mansfield	3.73E+03	5.63E+04	1.64E+05	2.24E+02
Bruce Mansfield	3.74E+03	3.82E+04	1.42E+05	1.84E+02
Bruce Mansfield	3.40E+03	2.51E+04	1.70E+05	2.00E+02

A.2 Information on Water bodies and Mercury Levels in Fish near the Bruce Mansfield Plant

Bruce Mansfield Power Plant, Shippingport, PA
From the Pennsylvania fish advisory 2004, there is only an advisory on PCBs.
No advisories for mercury in Beaver County.

From the EPA Fish database on mercury contamination levels (EPA, 1999):

Beaver County

Channel Catfish
Beaver River
Lat 40.7422 Long –80.3164, date: 95/10/04
Composite sample, n=5, Hg concentration = 0.070

Small mouth bass
Raccoon Creek
Lat 40.6278 Long –80.3378
Date: 95/09/26
Composite sample, n=5, Hg = 0.080

Common Carp (two samples)
Raccoon Creek
Lat 40.7442 Long -80.3164
Date: 95/10/04, composite sample n=5, Hg = 0.120

Large Mouth Bass
Travis Creek
Lat 40.5031 Long -80.4239
Date: 95/10/10, composite sample n=5, Hg = 0.460

Although the number of fish samples is small for each species, the values presented here are typical and do not provide cause for issuing fish advisories. Large mouth bass, which is the top-level predator among this group of fish have the highest Hg values.

A.3 Water bodies within 25 km of the Bruce Mansfield Power Plant

Table A-2. Water Bodies within 25 km of the Bruce Mansfield Plant.

Nearest Town	Water Body	Lat	Long	Hg
	Ohio River	40.67176	80.51766	
Larenceville	large unmarked res	40.62	80.51	
	Lake Ehs-Vel	40.67	80.68	
	Yellow Ck	40.61445	80.68	
	California Hollow Ck	40.343	80.56	
	Long Run	40.672	80.6306	
	Island Run	40.6855	80.517	
	Little Beaver Run	40.67176	80.53639	
	Upper Dry Run	40.6572	80.49855	

	Wolf Run	40.658	80.44178	
	Six Mile Run	40.68	80.4223	
	Six Mile Run	40.671	80.40	
	Ohio River	40.656	80.379	
	Moon Run	40.6707	80.2904	
	Crows Run	40.66	80.66954	
Freedom	Snake Run	40.19	80.22	
Freedom	Pine Run	40.19	80.20	
Freedom	Crows Run	40.68	80.2144	
Beaver	Two Mile Run	40.69956	80.29011	
Fairview	Bieler Run	40.70	80.4794	
Fredricktown	Little Beaver Ck	40.7004	80.536	
Fredricktown	Little Beaver Ck	40.714	80.555	
	Rough Run	40.729	80.593	
BrandyRumPark	South Branch Brandy Run	40.7287	80.355	
	North Branch Brandy Run	40.70	80.366	
	Two Mile Run	40.70	80.35	
Morade	Wallace Run	40.79	80.364	
Darlington	Beaver Ck.	40.801	80.443	
Darlington	Painter Ck.	40.786	80.46	
	Little Beaver Run	40.791	80.4787	
Cannelton	Bush Run	40.76	80.479	
E. Carmel	Spruce Lake	40.76	80.59246	
Mill Rock	Leslie Run	40.79	80.52	
Continue over				
Mill Rock	State Line Lake	40.83	80.516	
Negley	Leslie Run	40.8	80.54	
Darlington	Darlington Lake	40.82	80.486	
Homewood	Charles Run	40.81	80.364	
Homewood	Charles Run	40.786	80.3268	
Homewood	Beaver River	40.628	80.5176	
Glascow	Unmarked Res	40.628	80.5178	
Lawrance	Ohio River	40.51768	80.51768	
Glasgow	Upper Dry Run	40.65	80.498	
Glasgow	Lake Cha-Vel	40.67	80.687	
Hillcrest	Yellow Ck	40.62	80.669	
Hockstown	Mill Ck	40.61397	80.479	

Shippingport	Reggo Run	40.6138	80.47	
GreenGarden	Raccoon Ck	40.61328	80.3288	
Allquippa	Beaver River	40.61215	80.234	
	Ambridge Res	40.58446	80.3669	
Yellow Creek	Hollow Ck.	40.528	80.65	
Moscow	Tomlinson Run Lake	40.50	80.62	
	Tomlinson Run	40.556	80.542	
BonMeade	Flangherty Run	40.5264	80.2538	
	Swickley Cr.	40.569	80.178	
	Little Troueverse Ck.	40.52699	80.356	
Cumberland	Hardin Run	40.4988	80.556	
	Holbert Run	40.4699	80.5942	
New Summert	Hollow Rock Run	40.52796	80.669	
Imperial	McGlavens Run	40.4685	80.1284	
Bald Nob	Gorden Run	40.46874	80.30	
Rabinson	Bigger Run.	40.46915	80.349	
Cumberland	Holbert Run	40.4699	80.517	
Chestnot Hill	Kings Ck.	40.427	80.557	

Appendix B: Literature Review on Exposure and Modeled Deposition

B.1 Exposure

B.1.1 Fish Consumption

Newark NJ: Thirty percent or more of the people who fish and crab in Newark Bay Complex did not eat their self-caught fish or crabs. 8-25% of the people ate more than 1500 g/month. Some people angling in the Newark Bay Complex are eating crabs at a rate well over 1500 g/month and about 70% are eating crab and about 70% are eating crabs even though there is a total ban on both harvest and consumption. J. Burger. 2002. *Consumption patterns and why people fish. Environmental Research Section A 90*:125-135.

New Jersey: Fishing in urban New Jersey: Ethnicity affects information sources, perception, and compliance. People continue to eat their catch with a high percent of Hispanics and blacks. Twenty-six percent believe the fish are OK, 12% believe there is no risk to an unborn, and 25% are aware of warnings (J.Burger, K.K. Pflugh, L. Lurig, L.A. Von Hagen, S. Von Hagen. Fishing in Urban New Jersey. 1999. *Ethnicity Affects Information Sources, Perception and Compliance. Risk Analysis 19*:217-229).

Estimation of fish consumption and methyl mercury intake in the New Jersey population. *J. Exposure Analysis and Environmental Epidemiology 6:* 503-525. Of 1000 respondents, 933 reported that normally eat fish at least a few time/year. Six eighty six reported that consumed fish within the seven day recall period, yielding 177 fish meals or 2.1 fish meals/week per consumer (Stern, A.H., L. Korn, B.E. Ruppel. 1996. *J. of Exposure Analysis and Environmental Epidemiology* 6:503-525).

Savannah River: Factors in exposure assessment: ethnic and socioeconomic differences in fishing and consumption of caught fish in the Savannah River. Although most of those interviewed were men, they stated that wives and children ate fish as often ad they did. Children begin eating fish at 3-5 years of age. People fishing along the Savannah River at an average of 1.5 kg/month (J. Burger, W.L. Stephens, Jr., C.S. Boring, M. Kuklinski, J.W. Gibbons, M. Gochfeld 1999. *Risk Analysis 19*:427-437).

Lake Ontario: Communicating contaminant risks from sport-caught fish: the importance of target audience assessment. Fishing for food was not a target for any of these audiences, but all audiences at a portion of their catch and gave some to friends. Approximately 50% of migrants gave away some of their catch and 90% kept some to eat. Lake Ontario fishers were divided into groups based on the number of fish meals eaten in 1988: non-eaters 0, moderate eaters, 1-12, frequent eaters 13-52, high eaters greater than 52 (Velicer, C.M. and Knuth, B. 1994. Comunicating Contaminant Risks from Sport-Caught Fish: the Importance of Target Audience Assessment. *Risk Assessment 14*:833-841).

Identification of sport fish consumption patterns in families of recreational anglers through factor analysis. Environmental Research Section A 89: 19-28. Fish consumption patterns in families of recreational anglers. By five years of age, almost 50% of the children had consumed at least one

meal of sport fishing. A median of two to four sport fish meals were reported to be consumed by children for each year of life (Behler, G.P. 2002. Identification of Sport Fish Consumption Patterns in Families of Recreational Anglers through Factor Analysis. *Environmental Research Section A 89*:19-28).

Santa Monica Bay: In Santa Monica Bay. In 113 separate surveys 2,378 anglers were interviewed by mail. Pier and jetty anglers had lower household income than other anglers. Most anglers fish year round, but 19% only fished in summer. 77% of anglers were aware of health warnings. The surveys also included consumption information, Figures B-1 – B-3. The three figures denote the 50th and 90th percentile in the distribution for consumption (g/d). The 50th percentile is generally less than 25 g/d and the 90th percentile is less than 100 g/d. Seafood Consumption Habits of Recreational Anglers in Santa Monica Bay. Young et al., 1978, Gossett et al. 1983, Risebrough 1987, Pollock et al 1991, SCCWRP et al. 1992, SCCWRP 1994. (see Figure 1,2 and 3). http://www.sccwrp.org/pubs/annrpt/93-94/art06.htm

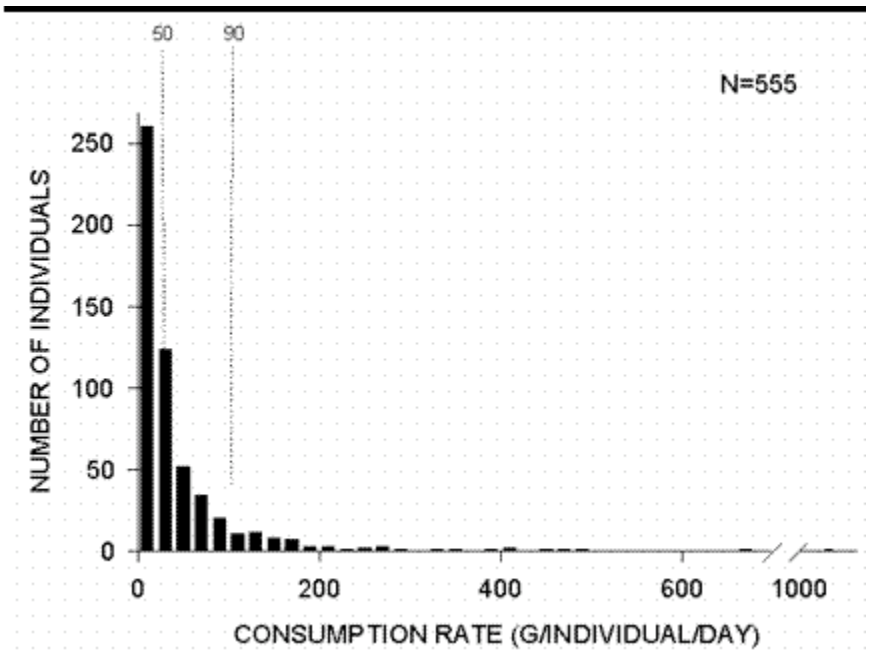

FIGURE B.1.
Distribution of consumption rates of all seafood species combined by ethnic groups of recreational anglers interviewed in Santa Monica Bay Seafood Consumption Study, September 1991 to August 1992. Median (50th percentile) and upper decile (90th percentile) are noted with vertical dotted lines.

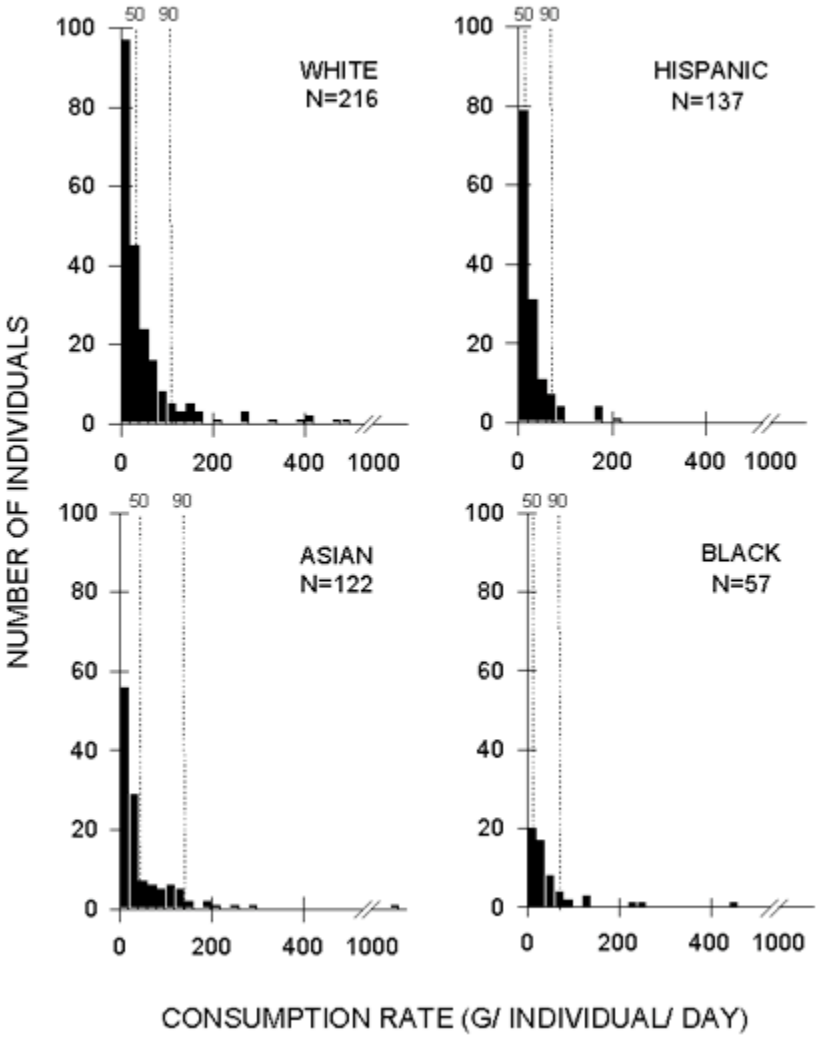

FIGURE B.2.
Distribution of consumption rates of all seafood species combined by annual household income groups of recreational anglers interviewed in Santa Monica Bay Seafood Consumption Study, September 1991 to August 1992. Median (50th percentile) and upper decile (90th percentile) are noted with vertical dotted lines.

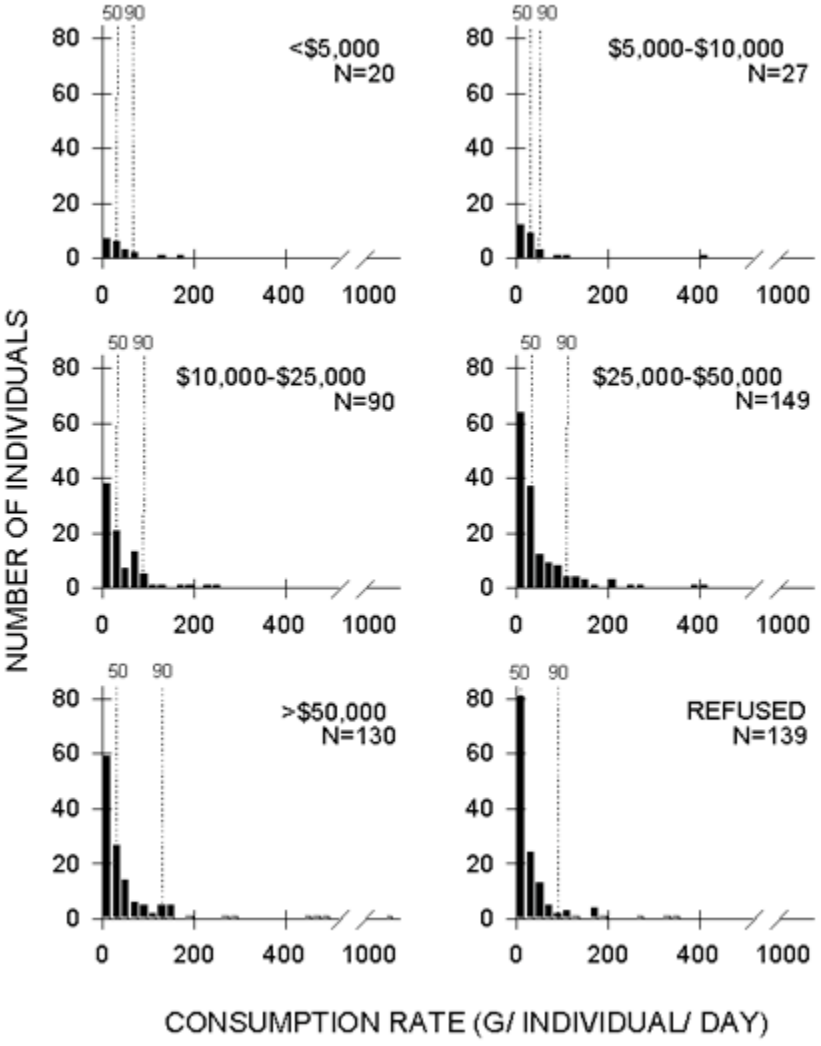

FIGURE B.3.
Seafood consumption rates by seafood species for recreational anglers interviewed in Santa Monica Bay Seafood Consumption Study, September 1991 to August 1992.

EPA Chemicals in fish: Consumption of fish and shellfish in California and the United States; Final Report. 2001

For groups of individuals who consume sport fish and/or shellfish, there is a continuum ranging from intermittent fishers, who may eat fish only occasionally, to those who fish regularly and/or heavily and consume large quantities of the fish that they catch. These "high-end consumers" could include recreational fishers with high rates of success and subsistence fishers who rely on their catch to feed themselves and their families. Overall national mean per capita consumption values for fish and shellfish combined ranged from 10 to 17.9 grams per day, excluding rates that were reported to be unreliable. Several studies have suggested that fish consumption rates differ for specific subpopulations. Demographic characteristics, such as race or ethnicity, age, and sex, and the relationships between these variables and fish and shellfish consumption rates and patterns are described below. Some of the national studies (*e.g.*, Rupp *et al.*, 1980) indicated regional differences in consumption rates that could be influenced by local cultural preferences and/or the types of fish and shellfish that are available in certain regions and at certain times of the year, and the amount of sport or subsistence fishing that occurs in a given region. In addition,

as indicated previously, rates may vary within the subset of the population that catches and consumes fish. Some recreational fishers may fish primarily in response to the seasonality of their "favorite" species, whereas other recreational fishers may fish more avidly. Subsistence fishers and recreational fishers may also preserve fish for consumption during non-fishing seasons or other times (Puffer *et al.*, 1982; U.S. EPA, 1996b). Subsistence fishers, by definition, are likely to fish on a regular basis in order to secure food for themselves and their families. Thus, subsistence fishers are typically considered to be high-end consumers. There are no particular criteria or thresholds that definitively describe the group. Additionally, fishers are not always willing to report their income and do not necessarily identify themselves as subsistence fishers. Thus, it may be difficult to define and represent subsistence fishers in a quantifiable way. Furthermore, definitions and perceptions of what constitutes "subsistence fishing" are likely to vary among regions and cultures.

Estimated Per Capita Fish Consumption in the United States August. 2002. Pesticide and Environmental Toxicology Section Office of Environmental Health Hazard Assessment California Environmental Protection Agency

The estimated mean daily average per capita consumption of "as prepared" freshwater and estuarine finfish and shellfish is 4.58 ± 0.42 grams/person/day. When consumption is estimated per kilogram of the consumer's body weight, the mean daily average of "as prepared" freshwater and estuarine finfish and shellfish consumption is 70.79 ± 6.23 milligrams/kilogram of body weight/day. These estimates project "as prepared" fish consumption data from a combined sample of 20,607 individuals to the population of 261,897,236 individuals in the 50 states and District of Columbia. A 90–percent confidence interval about the estimated national mean consumption of "as prepared" freshwater and estuarine finfish and shellfish is 4.16 to 5.00 grams/person/day, which indicates that there is a 90–percent statistical confidence that the true mean consumption is contained in this interval.

Section 4 of the report lists additional "as prepared" fish consumption estimates from the combined 1994-1996 and 1998 CSFII surveys. Included in these sections are point and interval estimates for upper percentiles from the empirical distribution of daily average per capita consumption of freshwater and estuarine species; mean and upper percentile estimates of marine species and all species; and estimates of finfish and shellfish consumption, both separately and together.

Section 4 also presents per capita fish consumption estimates for selected subpopulations in the United States. Some estimates extracted from this section include consumption estimates for women and low–income individuals.

Women aged 15 to 44 years, the childbearing years, consume a mean daily average of 4.28 grams of "as prepared" freshwater and estuarine finfish and shellfish. This same group consumed, on average, 7.03 ± 0.91 grams/person/day of "as prepared" marine finfish and shellfish.

Individuals in the low–income group, defined in the CSFII as individuals from households with gross incomes at or below 130 percent of the federal poverty threshold, consume a mean daily average of 11.32 ± 1.67 grams/person/day of "as prepared" finfish and shellfish. The estimated mean daily average consumption of "as prepared" freshwater and estuarine finfish and shellfish by low–income individuals is 4.35 ± 0.92 grams/person/day.

Information on substantial fish consumption groups

CHEMICALS IN FISH: CONSUMPTION OF FISH AND SHELLFISH IN CALIFORNIA AND THE UNITED STATES FINAL REPORT October 2001 Pesticide and Environmental Toxicology Section Office of Environmental Health Hazard Assessment California Environmental Protection Agency

U.S. EPA (1994, 1996) considered subsistence fishers to be people who rely on noncommercial fish as a major source of protein, and suggested that subsistence fishers tend to consume noncommercial fish and/or shellfish at higher rates than other fishing populations, and for a greater percentage of the year, due to cultural and/or economic factors. However, EPA (1996) also noted that consumption rates can vary considerably among subsistence fishers. Few studies have specifically targeted fishing populations identified as subsistence fishers although U.S. EPA (1996b) indicated that several studies were in progress or recently completed. The definition for subsistence fishers provided by U.S. EPA is a narrative and does not indicate how to actually identify subsistence fishers in a population. There are no particular criteria or thresholds (such as income level or frequency of fishing) that definitively describe the group.

Additionally, fishers are not always willing to report their income and do not necessarily identify themselves as subsistence fishers. In some of the "subsistence studies" summarized by U.S. EPA (1996b), the respondents indicated that they did not consider themselves subsistence fishers although they relied on the fish they caught as a major component of their diet.

Other subpopulations may be considered to be subsistence populations even though their reported rates of fish consumption are similar to what has been reported nationally for the U.S. population (*e.g.*, APEN, 1998; Shubat *et al.*, 1996). Thus, it may be difficult to define and represent subsistence fishers in a quantifiable way. Furthermore, definitions and perceptions of what constitutes "subsistence fishing" are likely to vary among regions and cultures. Some examples of differences in fish consumption rates reported for "subsistence" populations in different regions follow in the discussion below.

U.S. EPA (1997c) suggested that Native American, lower income urban, rural, and Asian-American populations often include subsistence fishers, and described some of the difficulties in characterizing these subpopulations in general, and subsistence fishers, in particular. For example, subsistence fishers may not have registered for fishing licenses for a variety of reasons, and thus are likely to be underrepresented in surveys based on fishing licenses. In addition, U.S. EPA (1996b) noted that fish consumers might answer survey questions inaccurately for a number of reasons, including language problems, pride, concerns about illegal activity, and fear of restrictions that might jeopardize the fisher's family and/or access to fishing resources.

Problems with defining subsistence fishers contribute to difficulties characterizing consumption of fish and shellfish by the subpopulation of subsistence fishers. The Santa Monica Bay study asked respondents to report their annual household income, Figure B.3. Relatively few respondents reported annual household income in the lowest income brackets (less than $5000 or between $5000 and $10,000) and it is not possible to determine whether any, some, or all of these people, or those with higher income, were subsistence fishers. Furthermore, although the median rate of consumption was higher for the lowest-income group, the mean and upper percentile rates were higher for the higher-income groups. The study also asked fishers about their frequency of fishing and found that roughly two percent of the respondents reported having fished every day for the prior month. However, a consumption rate was not determined separately for this group. Additionally, although subsistence fishers would be expected to fish frequently, other fishers (*e.g.*, retired persons) may also fish frequently. On the other hand, if at least some subsistence fishers were included in this survey, then the upper percentile consumption rates that were calculated would have represented this subpopulation to some extent.

B.1.2 Mercury in Tuna

Burger J, Gochfeld M. 2004. Mercury in canned tuna: white versus light and temporal variation. *Environ. Research 96*:239-249, burger@biology.rutgers.edu

There are abundant data and advisories for mercury levels in wild fish, but far fewer for commercial fish that compose a large majority of the fish most people eat. Until recently, relatively little attention has been devoted to examining mercury in canned tuna, despite its great importance in human diets. There is substantial media coverage of the benefits and risk from fish consumption, but few peer-reviewed data on canned tuna, the most commonly consumed fish in the United States. In this paper, we examine the levels of total mercury in canned tuna obtained from a New Jersey grocery store from 1998 to 2003, looking for temporal consistency within this data set and particularly for comparison with the Food and Drug Administration's 1991 study. We analyzed 168 cans individually for total mercury. All values are reported as parts per million (= microg/g) on a wet weight basis. In a subset of samples analyzed for total and inorganic mercury, the inorganic mercury was below detection levels; hence at least 89% of the mercury can be considered methyl mercury. We found that white-style tuna had significantly more total mercury (mean 0.407 ppm) than light-style tuna (mean 0.118 ppm), presumably reflecting that "white" tuna is albacore, a species relatively larger than the skipjack tuna, which is commonly available as "light" or "chunk light." The maximum mercury in a can was 0.997 ppm, but 25% of white tuna samples exceeded 0.5 ppm. Data suggest a slight increase in levels since 1991, and mercury levels were significantly higher in 2001 than in other years. The mean level of mercury in white tuna (mean 0.407 ppm) was significantly higher than the mean value of 0.17 ppm currently used by the U.S. Food and Drug Administration (FDA) in its risk assessment and public information. There were no significant differences in mercury levels in tuna packed in oil compared to water. Draining contents had no effect on mercury levels, and the fluid, both oil and water, contained little mercury. These data indicate that people who eat canned tuna frequently can choose light tuna and reduce their mercury intake. Canned mackerel had much lower levels of mercury than tuna. Since cans of white tuna frequently exceed the FDA's original action level of 0.5 ppm, it would be prudent to continue some systematic monitoring of the nation's canned fish supply, particularly as the targets of commercial fisheries inevitably change as certain stocks become depleted.

B.2 Modeled Deposition

Comparison of Modeled Depositions under MACT or Cap and Trade Regulations (EPRI, 2004)

EPRI has applied state-of-the-art modeling to evaluate the potential for hot spots under alternative approaches to regulating utility mercury proposed by EPA. Because it is impractical to look for “hot spots” by measuring mercury deposition at every location in the country, EPRI has run sophisticated, state-of-the-art computer models to simulate deposition of the mercury released from power plants and other emission sources. EPRI’s analysis considered the amount and chemical forms of mercury emitted from every coal fired power plant in the United States under three scenarios: a 2004 Base Case, for current conditions; and EPA’s two proposed regulatory approaches, the MACT rule and the Cap & Trade rule (EPRI, 2004).

The model simulations of regulatory scenarios are for year 2020, when all emission reduction measures mandated by either rule will be fully implemented. The Base Case simulates mercury

emissions from power plants and all other mercury sources, such as municipal and medical waste incinerators. The two regulatory scenarios lower power plant emissions according to requirements of the proposed MACT or Cap & Trade rules, but keep emissions from other sources constant. Holding emissions from other sources constant while varying emissions only from power plants allows researchers to estimate the impacts of EPA's proposed approaches to regulating utility mercury. To perform the simulations, EPRI used a national economic model to evaluate the amount and chemical forms of mercury emitted from U.S. power plants under each scenario. These emission results were fed into a fine-scale model of mercury chemistry and physics in the atmosphere, which was used to calculate amounts and patterns of deposition throughout the United States under current conditions, the MACT rule, and the Cap & Trade rule (EPRI, 2004).

EPRI's results show that the highest values of modeled deposition in the United States are produced by mercury emitted from sources other than power plants. According to EPRI's computer simulations, *after regulation*, the areas of highest mercury deposition in the United States will continue to be those locations chiefly affected by emissions from sources other than power plants (EPRI, 2004).

Even with a liberal definition of utility-influenced deposition locations (i.e., where utility-emitted mercury makes up roughly 30% or more of the total deposition), only about 2.5% of US surface area falls into this category following MACT or Cap and Trade (EPRI, 2004).

The "Hot Spots" in mercury deposition predicted by modeling suggest that these locations receive most of their mercury from municipal and medical waste incinerators. Locations affected by these incinerators would continue as the leading areas of mercury deposition in the mid-Atlantic and southern New England states, even after power plants have fully reduced their emissions. This result holds for either the MACT rule or the Cap & Trade rule. While distant non-U.S. mercury sources are the dominant contributors to deposition in much of the United States, non-utility U.S. sources emit mercury at rates and in forms that dominate deposition in their regions. Even after power plant sources are controlled, incinerators will continue to dominate the mercury contributing to deposition in high-deposition areas. Both regulatory approaches proposed by EPA would play an important role in reducing deposition in locations that have substantial deposits from utility sources in 2004 (EPRI, 2004).

After full implementation of MACT or Cap and Trade regulations, locations where depositions originating from power plants are dominant will fall below the top 55 locations of highest human-caused deposition. However the Cap and Trade approach would produce markedly lower deposition at utility-dominated locations than would the MACT approach. Neither proposed regulatory approach would increase deposition in high-deposition areas or create new high-deposition areas compared to current levels (EPRI, 2004).

The Cap and Trade rule produces greater mercury deposition reductions than does the MACT rule. Modeling results for 2020 show that all states in the country will experience overall reductions in deposition due to the proposed mercury rules. But the reduction in mercury deposition is greater under the Cap & Trade rule (an average drop of 7%) than under the MACT rule (an average drop of 5%). Reductions in deposition vary somewhat by location, with greater reductions occurring in the mid-Atlantic and Southeastern states where a higher fraction of bituminous coal is burned. Bituminous coal emits a relatively higher proportion of divalent mercury— the chemical form most easily captured by currently available NOx and SOx emission control devices, as well as by mercury-specific control devices currently under

development. Since it is more cost-effective to reduce mercury emissions at these plants, they are more likely to install controls and therefore will have a greater relative impact on reducing mercury emissions and deposition (EPRI, 2004).

For many air pollutants, the concept of a “hot spot" is associated with higher levels of inhalation risk due to high concentrations of the pollutant in the atmosphere. Evidence shows there is no cause for concern about inhalation risk due to atmospheric concentrations of mercury. The EPA threshold of health concern for long-term inhalation exposure to mercury is expressed as an atmospheric concentration of 300 ng/m^3. This means that a person can inhale mercury at this concentration for a lifetime with no appreciable risk of ill effects. Even for 2004 levels of mercury emissions, prior to new utility mercury controls, the highest modeled annual average atmospheric concentration is only 7.4 ng/m^3, more than 30 times less than the EPA health threshold. This worst-case current exposure drops even more under either regulatory approach, so that mercury exposure by inhalation will not be a public health concern at any U.S location.

Appendix C: Annotated Literature Review on the Perception of Mercury Risks

The Impacts of Recent Health and Environmental Research on Perceptions of Mercury Risks from Coal-Fired Power Plants

Fred Lipfert
Independent Consultant
Northport, New York

Terry Sullivan
Brookhaven National Laboratory
Upton, New York

An Annotated Compendium of Recent Publications on Mercury Issues, Presented At the Western Fuels Symposium, 19th International Conference on Lignite, Brown, and Subbituminous Coals, Billings, Montana, October 12, 2004

C.1 Health Effect News

Study of 212 US Preschool Children Finds PCBs More Important than Mercury in Terms of Cognitive Development

Paul Stewart, Jacqueline Reihman, Edward Lonky, Thomas Darville, and James Pagano

Oswego, New York, January 2003. The effects of fish contamination were investigated by comparing the test performances of 212 children whose mothers ate varying amounts of local fish during pregnancy. Great Lakes fish are contaminated with Pb, PCBs, and methyl mercury (MeHg). Thus study compared the development of children of 152 women who never ate Great Lakes fish with those of 141 women who had eaten a total of at least 40 lbs. The McCarthy Scales of Children's Abilities were administered when the children were 38 months old and again at 54 months. The fish contaminants considered included PCBs and lead, as measured in the umbilical cord blood, and methyl mercury (MeHg), as measured in the mothers' hair. The statistical analysis of the first test results showed a significant decrease in test performance with increasing levels of the most highly chlorinated PCBs, with or without adjustment for confounders (MeHg); mercury exposure was only weakly correlated with PCB exposure. In regression analyses using MeHg as the independent variable, with the cohort stratified by PCB exposure, MeHg was not a significant predictor of test performance for the entire group or for those with low PCBs. However, the interaction between MeHg and PCBs (high exposures of both) was significant at 38 months but not at 54 months. PCBs (alone) were not significant in the later tests, suggesting functional recovery with age. No relationships were found between test performance and fish consumption *per se* (average < 15 meals/yr), confirming the absence of MeHg effects and suggesting that other food sources of PCBs may have been important.

Published in Neurotoxicology and Teratology, Volume 25, pp 11-22, 2003.

Our Comments: The average Hg content of the hair of these mothers (0.5 ppm) is similar to the mean of a national sample of those eating fish three or more a times per month (see Figure C-1). Thus, this cohort study is one of the first that is directly applicable to the United States. The study shows no adverse effects of Hg per se, but finds that there may be an interactive effect when Hg and PCB exposures are combined. These exposures were combined in the Faeroe Islands Study, but Seychelles study was free from PCBs. The overall message is that, not only will reducing Hg emissions in the U.S. have negligible benefits to public health, local fresh-water freshwater fish may not be entirely safe to eat until PCBs are eliminated from our diet.

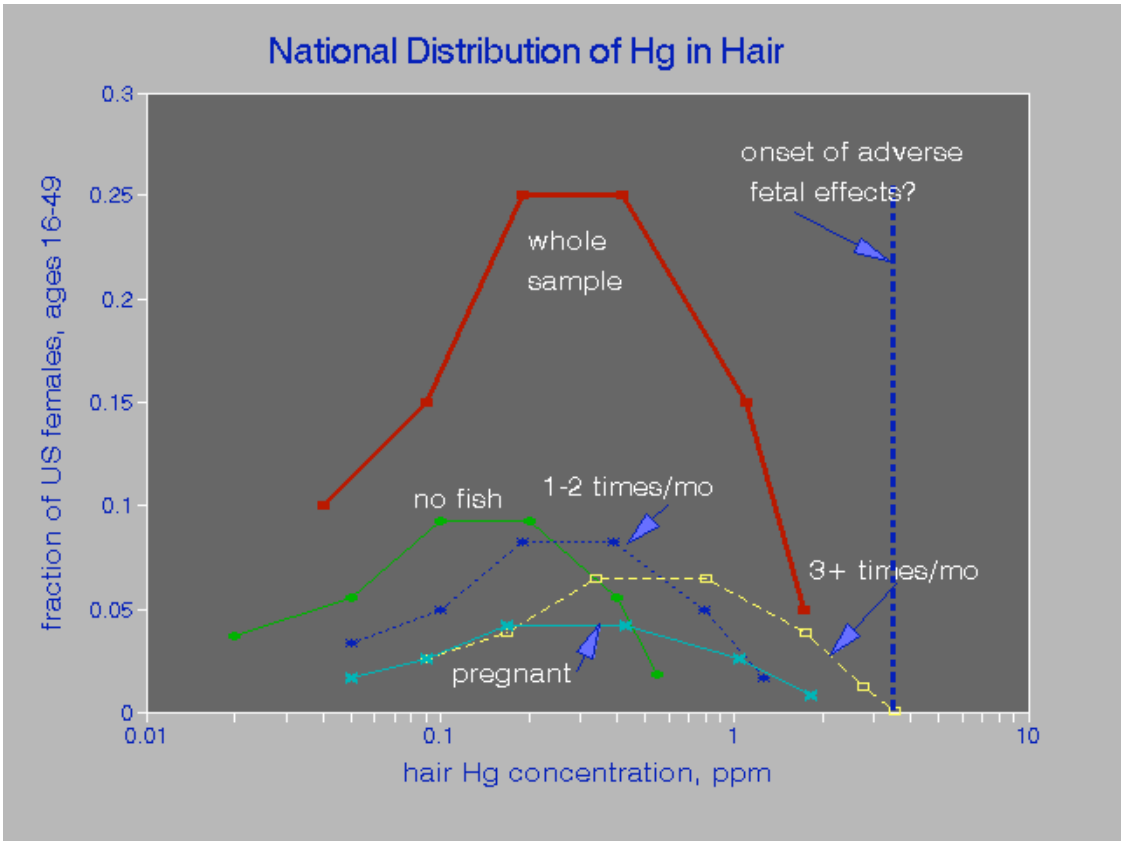

Figure C-1. National distribution of mercury in women's hair. Source: NHANES data, 2003.

Infants Whose Mothers Ate More Fish During Pregnancy Show Enhanced Development, Mercury Content Notwithstanding

Julie Daniels, Mathew Longnecker, Andrew Rowland, and Jean Golding

Bristol, England, March 2004. The cognitive development of 7,421 British children was tested using the MacArthur Communicative Development Inventory at age 15 months and the Denver Developmental Screening Test at 18 months. According to the questionnaires they returned, 39% of these mothers ate fish 4 or more times per week while pregnant; 31%, 1-3 times, and 30% either less than once per week or never. Mercury exposure was measured in a subset of 1054 children, in terms of the content of umbilical cord tissue. The statistical analysis adjusted for sex, birth order, maternal age and education, breastfeeding status, dental treatment, smoking and alcohol use during pregnancy, fish intake at other times, the child's age at testing, and the home environment. Higher fish consumption was associated with higher mercury exposure levels but with higher development test scores; the differences were small but statistically significant. There was no statistical relationship between mercury exposure and child development, but Hg exposures were much lower than in the Faeroe Islands, where adverse effects have been found.

Published in Epidemiology, Volume 15, pp. 394-402, July 2004

Our Comments: Although the mercury exposures in this cohort are only about 13% of those in the Faeroes, from which data were derived for the EPA reference dose, they are nevertheless

about three times the U.S. national average for women of childbearing age. American women eat much less fish than the members of this cohort, which is line with the difference in mercury exposures. By implication, the U.S. population is not being exposed to harmful levels of mercury. ***Fish is a good brain food after all!***

American Academy of Pediatrics Assesses Effects of Environmental Chemicals on the Developing Embryo, Infant, Child, and Adolescent

The Pediatric Perspective: Robert Brent, Suzanne Tanski, and Michael Weitzman

Wilmington, Delaware, October 2003. Because the embryo and the child are growing, adverse effects may occur at lower exposures to some chemicals, drugs, and physical agents. However, children and adolescents have better recuperative capacities than adults for many toxic agents. Discussions of the toxicity of environmental agents tend to provoke controversy. Segments of the scientific and lay communities tend to believe that environmental agents are major hazards, while others believe that these risks have been grossly exaggerated. Some scientists suggest that the permissible exposures for children should be cut by a factor of 10 to allow for uncertainties, but there is little scientific evidence to support this contention. Only the facts, impeccable science, and more research will put environmental effects on children into proper perspective. Single studies do not refute or demonstra9te causality.

Mercury Exposure and Child Development: Philip Davidson, Gary Myers, and Bernard Weiss

Rochester, New York, October 2003. The Seychelles and Faeroe Island studies of child development and mercury exposure came to different conclusions: no adverse effects in the Seychelles, and adverse associations with various developmental measures in the Faeroes. Both studies have been judged methodologically sound and scientifically valid. There are other differences between the cohorts, chiefly the exposure to PCBs in the Faeroes from eating whale. The Faeroes investigators reported that mercury neurotoxicity might be exacerbated by PCBs, but they continue to believe that direct adverse effects of Hg are present. EPA used these data to develop their "reference dose" for mercury exposure. Part of the result has been fear of eating fish, by some consumers.

Published in a Special Issue of Pediatrics, Volume 113, April 2004, Part 2. 240 pp.

Health Was Not a Major Focus of the 7th International Conference on Mercury as a Global Pollutant (ICMGP)

Llubljana, Slovenia, June 27-July 2, 2004. The program for the seventh in this series of international conferences comprises 630 papers and posters, of which only four were concerned with the epidemiology of consuming fish contaminated with methyl mercury. There were a few papers on fish-related exposure and on toxicology and the effects of dental amalgams, but the bulk of the conference was concerned with emissions, analytical methods, ecosystem effects, and atmospheric processes. The information below is based largely on conference abstracts, together with related prior publications.

The Seychelles Child Development Study

Recent analyses of prenatal MeHg exposure.

Conrad Shamlaye, Gary Myers, Philip Davidson, Christopher Cox, Li-Shan Huang, Thomas Clarkson

Rochester, New York, June 2004. The Seychelles children have been evaluated six times, through 11 years of age, and no consistent patterns of adverse associations have been found using a linear dose-response function. However, nonlinear models suggest subtle developmental changes at high exposures (hair Hg > 10 ppm). Previous analyses at 66 months of age produced conflicting non-linear results.

Analyses of postnatal MeHg exposure.

Gary Myers, Sally Thurston, Conrad Shamlaye, Philip Davidson, Christopher Cox, Thomas Clarkson

Rochester, New York, June 2004. Convenience samples of the children's hair have been obtained at several intervals and analyzed for postnatal MeHg exposure. Evidence from animal studies suggests there may be latent effects from postnatal exposure. Based on hair Hg and testing at 66 months, there was a decline in test performance for children's hair Hg levels above 10 ppm, which is well above levels experienced in the U.S.

Our Comments: The median hair mercury content of the Seychelles mothers was 6.9 ppm and they ate ocean fish 12 times per week, on average. These exposures are about 30 times the U.S. average, which would appear to yield an adequate factor of safety for the U.S. population.

Children's Health and the Environment in the Faeroes

Esben Budtz-Jorgensen and Philippe Grandjean

Copenhagen, Denmark, June 2004. Three different exposure measures (umbilical cord blood Hg, maternal hair Hg, frequency of eating whale) were compared using structural equations and factor analysis. All three measures were transformed to logarithms, because of the skewed distribution of cord blood Hg (covering a factor of ~300). The lack of agreement among these measures is interpreted as evidence of exposure error, ranging from 56-60% (2 standard deviations) for cord blood, 104-110% for hair, and even more for consumption of whale. These

uncertainties are much larger than those indicated by laboratory error and indicate that the true dose-response functions (based on logarithms) may be even steeper than previously reported.

The brainstem as a target of developmental methyl mercury toxicity.

Philippe Grandjean, Katsuyuki Murata, Esben Budtz-Jorgensen, and Pal Weihe

Copenhagen, Denmark, June 2004. Experimental studies suggest that the brainstem (the part of the brain connecting to the spinal cord) may be a sensitive target for MeHg. Brainstem auditory evoked potentials (BAEPs) were determined by presenting click signals to the right ear, masking the other ear with white noise, and measuring electrical response signals with brain electrodes. Heart-rate variability was also measured and was correlated with BAEPs. The results suggest a neurotoxic mechanism involved in cardiovascular abnormalities and increased mortality related to dietary MeHg exposure.

Our Comments: Exposure misclassification or measurement error has other statistical ramifications. For example, when a regression slope is biased low, the x-axis intercept is shifted to the left, thus obscuring any thresholds that might be present. When two competing risk factors, such as MeHg and PCBs, have different degrees of measurement error, the one with the least error tends to prevail. So far, PCB exposure errors have not been specifically addressed, but only 3 PCB congeners were considered and only for about half of the children. BAEPs are subclinical indications of response and their significance to childhood development is unclear. Only 1 of the 20 determinations based on children's hair Hg was significant, thus casting doubt on the claim of irreversible effects. The responses to maternal hair suggested an increase above 10 ppm.

OUR BOTTOM LINE

The dichotomy between the two major studies of the outcomes of prenatal exposures (Seychelles vs. Faeroes) is unlikely to be resolved and should be declared a "standoff". There are important differences in the populations, their diets and exposures to other toxins (PCBs), the measures of neurological outcome, and the statistical methods used. Instead, more attention should be given to studies of the U.S. population, and a new large-scale study should be considered. The extant exposure studies suggest that it should be possible to compare neurological outcomes in sufficiently large numbers of mothers having exposures above and below the present EPA guideline. Such a study should include complete dietary patterns and other neurotoxins (PCBs). The objectives of such a U.S.-based epidemiological study would be to develop relationships for use in risk analyses, not necessarily to develop alternative public health guidelines.

C.2 Exposures to Methyl mercury

National Survey of Mercury Exposures Finds Low Values for U.S. Women of Childbearing Age, Based on Hg Content of Hair

Margaret McDowell, Charles Dillon, John Osterloh, Michael Bolger, Edo Pelizzari, Reshan Fernando, Ruben de Oca, Susan Schober, Thomas Sinks, Robert Jones, Kathryn Mahaffey

Hyattsville, MD, May 2004. Hair samples obtained from 1726 women aged 16-49 and 838 children aged 1-5 show mercury concentrations well below those at which adverse effects have actually been observed (see Figure C-1). The median levels in children and women were 0.12 and 0.20 ppm, respectively. This is consistent with the low rates of fish consumption that were reported; 39% of the women reported eating no fish during the past 30 days, 34% reported eating 1 or 2 fish meals, and only 27% reported eating 3 or more fish meals during that period. By extrapolation, the 99th percentile of all females would have a hair Hg content of about 5 ppm, and the 99th percentile of the fraction of all females who eat fish 3 or more times per month would have about 8 ppm. The 99th percentile of those who eat no fish would be about 1.3 ppm. The hair Hg values for women are well correlated with the corresponding concentrations in blood; the log-log correlation is 0.79 and the hair/blood slope is 234. Using these values, the maximum hair concentration in this sample would be about 9 ppm.

Published in Environmental Health Perspectives, Volume 112, pp. 1165-1171, 2004.

Our Comments: Median hair Hg levels in women of childbearing age have declined by 50% since 1981. However, the distribution has broadened; based on 2820 observations, the maximum level in 1981 was 6.9 ppm, and the apparent effect of seafood consumption has increased slightly. The interpretation of acceptable levels of MeHg exposure levels depends on the definition of "safe." EPA set their reference exposure level at 1.1 ppm, by imposing a factor of 10 "safety factor" on the derived benchmark value of 11 ppm. The Food and Drug Administration uses an advisory level of about 4.5 ppm, and other authors have set 3.5 ppm (shown on the graph) as the "onset of adverse effects." It is important to note the difference between a limit that has been set to ensure public health (like the speed limits posted for highway curves) and the levels at which even very subtle adverse effects have been observed. Note that the Clean Air Act was never intended to provide absolute protection to the entire population; if EPA's approach to mercury regulation were to be applied to all air pollutants, no area of the country could be in compliance.

Survey of American Diets Shows that Tuna is Common, but Fish that are High in Mercury Are Rare

National Center for Health Statistics, Centers for Disease Control

Hyattsville, MD. A 1999-2000 survey of about 10,000 subjects provided data on the rates at which various types of fish and shellfish are consumed, based on personal recall of the past 30 days. The table below gives the percentage of respondents reporting consumption of selected species and the median and maximum numbers of meals in 30 days. The maximum usually refers to a single respondent, or about the 99.99th percentile of the entire sample.

Species	**% of sample**	**median**	**max meals/month**	**Species**	**% of sample**	**median**	**max meals/mo**
all shellfish	39.7			pollock	1.8	1	12
all fish	60.5			haddock	1.8	1	8
tuna	28.3	1.5	50	bass	1.2	1	7
breaded products	13.5	1	28	swordfish	0.9	1	4
salmon	9.9	1	44	mackerel	0.6	1	8
catfish	8.8	1	15	porgy	0.4	1	10
cod	4.4	1	30	sea bass	0.5	1	7
flatfish	3.8	1	20	walleye	0.4	1	8
sardines	2.4	1	20	shark	0.3	0.5	6
perch	2.3	1	30	pike	0.1	1	8
trout	1.9	1	8	other fish	10.0	1	30
unknown fish	4.5	1	90				

Our Comments: These survey data show that tuna accounts for about half of all fish meals and probably more than half of the dietary intake of Hg. For most fish species, a total of about 2 meals were eaten per month, on average (2.5 for tuna and "other"). Predatory and fresh-water fish are only minor contributors to the U.S. diet. It is perhaps apocryphal that the one respondent who reported eating fish at every meal didn't know what kind it was!

C.3 Effects Of Nutrition

Most of the epidemiology studies on methyl mercury exposure from eating fish neglect any effects from the subjects' overall diets. Although it was noted that the Iraqi population that ate seed grain that had been treated with mercury fungicide in 1971-2 was facing famine, the overall nutritional situations of the various populations under study have not been considered in detail. However, common sense shows that in any given situation, the only way to be highly exposed in the long term is to eat fish more often, since the Hg content of the fish involved will tend to average out. In the New Zealand study, those most highly exposed were said to be eating take-out fish-and-chips five nights a week; one must thus wonder which important dietary nutrients might have been missing from their diets. The questions of direct and indirect effects of diet and nutrition have now been raised in several papers.

Can Nutrition Affect Chemical Toxicity?

Arthur Furst (International Journal of Toxicology, Volume 21, pp. 419-424 (2002)).

San Francisco, CA, June 2002. This essay considers three examples. Selenium is shown to inhibit methyl mercury toxicity, especially with regard to aquatic processes. Oxidative stress is suspected as promoting some cancers, atherosclerosis, and the adverse effects of aging. Dietary anti-oxidants, such as fruits and vegetables, may thus be protective. Vitamin C can inhibit the conversion of nitrites in preserved meats to nitrosamines. However, there are large gaps about these processes in the existing knowledge base.

The Influence of Nutrition on Methyl mercury Intoxication

Laurie Chapman and Ling Chan (Environmental Health Perspectives Supplements, Volume 108, pp. 29-56 (2000))

McGill University, Quebec, Canada. This review comprises 321 references and discusses effects on pharmacokinetics, toxicity, and mechanisms. It concludes that a wide variety of foods and nutrients alter MeHg metabolism, but that the mechanisms of such interactions remain speculative. It seems clear that dietary factors need to be better addressed in future epidemiology and clinical studies.

Nutritional Factors May Modify the Toxic Action of Methyl Mercury in Fish-Eating Populations.

Thomas Clarkson and James Strain (Journal of Nutrition, Volume 133, pp. 1539S-1545S (2003)).

Berkeley, CA, June 2002. At an international conference on "Trace Elements in Man and Animals", the toxic properties of MeHg were reviewed and the potential role of micronutrients in fish was discussed. It was hypothesized that they might explain the lack of adverse (and often beneficial) effects of fish in the Seychelles. The compounds of interest include long-chain polyunsaturated fatty acids, iron, and chorine, which may protect against neurotoxicity.

Eating tropical fruit reduces mercury exposure from fish consumption in the Brazilian Amazon.

Carlos Passos, Donna Mergler, Elizete Gaspar, Silmara Morais, Marc Lucotte, Fabrice Larribe, Robert Davidson, Sylvia de Grosbois. (Environmental Research, Volume 93, pp. 123-130 (2003); Volume 96, pp. 102-105 (2004)).

Montreal, Canada, January 2003. A small group of female villagers (n=26) were studied for one year, including daily food diaries and monthly hair samples for Hg analysis. The results showed an inverse relationship between beef meals and hair Hg (the effect of substitution for fish) and an inverse relationship between hair Hg and meals including fruit (which was uncorrelated with fish meals). A stratified regression analysis showed that the effect of eating fish on hair Hg was about 5 times stronger in subjects who ate less fruit.

Our Comments: These papers consider three different aspects of the nutritional question: (1) direct interaction with the mechanisms of MeHg toxicity, (2) other effects of nutrients in fish, (3) interactions with MeHg exposure, perhaps with respect to absorption and excretion. Together with the effects of other toxins in fish, such as PCBs or Pb, the overarching message is that we should not expect to find agreement among disparate cohort studies conducted in very different parts of the world. The stark contrast between the Faeroes and Seychelles studies may be the best evidence of this precept. This reinforces the need for an American epidemiological study.

OUR BOTTOM LINE:

It is important to distinguish the exposures of the most susceptible subpopulations. Exposures of the U.S. general public, including pregnant females, have decreased in recent decades and are not a current concern, even though a small percentage may exceed EPA's public health guideline (1.1 ppm in hair). There is no evidence that exposures high enough to cause harm are being exceeded in the U.S., even by the pregnant women in the San Francisco study of "high-end" consumers of predatory fish. An alternative approach to risk analysis could involve comparing human exposures by proximity to concentrations of coal-fired power plants and other sources of atmospheric mercury.

C.4 Atmospheric Modeling and Ground Truth Data

Any projected health benefits from reduced power plant mercury emissions are based entirely on atmospheric modeling and theoretical rates of methylation and uptake by fish. Much of the world will agree that reducing global airborne mercury is a desirable goal. But, "the devil is in the details." Which sources of Hg are the most important? How much intercontinental transport is there? What are the spatial and temporal relationships between Hg deposition, fish Hg content, and Hg exposures of the most susceptible populations? Lacking such detailed information, assumptions are often the only recourse. Key atmospheric issues include:

- Accuracy and precision of large-scale atmospheric models.
- Validation of models against empirical data (the "Pennsylvania anomaly").
- Evidence for reduction of reactive gaseous mercury (RGM) to elemental Hg (Hg0) in power plant plumes.
- Relative importance of urban sources.

Global Emissions and Transport: What Is Known and Unknown.

Russell Bullock, Jr. (presented at the USGS/EPA Mercury Roundtable, Sept. 15, 2004).

Reston, VA, Sept. 15 2004. This paper presents some back-of-the-envelope estimates of the fraction of Hg deposited in the U.S. that originated elsewhere. It assumes that most of the imported Hg is elemental and that most of the Hg that deposits from any source is RGM, of which the U.S. emits about 50 metric tons per year (mt/y). US Hg emissions are only about 1.5% of the imported mass flux across the western boundary, which is why ambient Hg_0 is relatively uniform across the U.S. (and the globe). Chemistry (conversion of Hg_0 to RGM) and subsequent deposition thus controls the relative importance of sources. With a range of half-lives for Hg_0, the fraction of domestic deposition varies from 26% to 55%. However, EPA modeling shows hot-spots as high as ~80% in the northeast.

Our Comments*:* Measured rates of Hg deposition range around 10-20 µg/m2/y, which sums to 90-180 mt/y, compared with only 50 mt/y of RGM emitted in the U.S. This also corresponds to 28-56% deposition from U.S sources, of which the utility share is even lower, but the rate of conversion of Hg_0 to RGM appears to be key.

Review of Published Ground-Truth Data Shows Weak Local and Regional Effects of Mercury Emissions from Power Plants (also Chapter 2 of this report).

Fred Lipfert, Terry Sullivan, and Scott Renninger

Anaheim, California, March 29, 2004. Seven experimental studies of local Hg deposition and related effects were reviewed and reanalyzed, some dating back to 1973. Local effects (< 30 km downwind) indicated that only about 5-10% of emitted Hg was deposited and retained in soil or sediment cores. About 12% excess wet deposition was seen near one power plant. Some of the inferred deposition patterns suggest peaks close to the source (wet deposition) but also secondary

peaks downwind (dry deposition?). However, confounding by varying background levels could not be ruled out. Fish Hg was only weakly associated with estimated local deposition. At the regional scale, measured wet deposition (MDN) data were used to assess power plant impacts in Pennsylvania, where measured values are much lower than EPA predictions (the "Pennsylvania anomaly"). A weak relationship was seen with emissions, implying that a 50% cut would reduce wet deposition by only about 8%. At the national scale, state-level data on fish Hg content were examined in relation to state-wide average wet deposition. A weak, non-significant relationship was found, such that an 8% drop in wet Hg deposition would lead to a 1.7% decrease in fish Hg (see Figure C-2. Moreover, the Midwestern coal-burning states had among the lowest wet deposition rates and fish Hg levels. One of the conclusions is that local excess deposition seems to exist around power plants, but fish Hg content is not always increased, probably because of local differences in watersheds and methylation rates.

Presented at the 227th National Meeting of the American Chemical Society

Our Comment: Given the uncertainties in atmospheric modeling, additional ground truth data are badly needed.

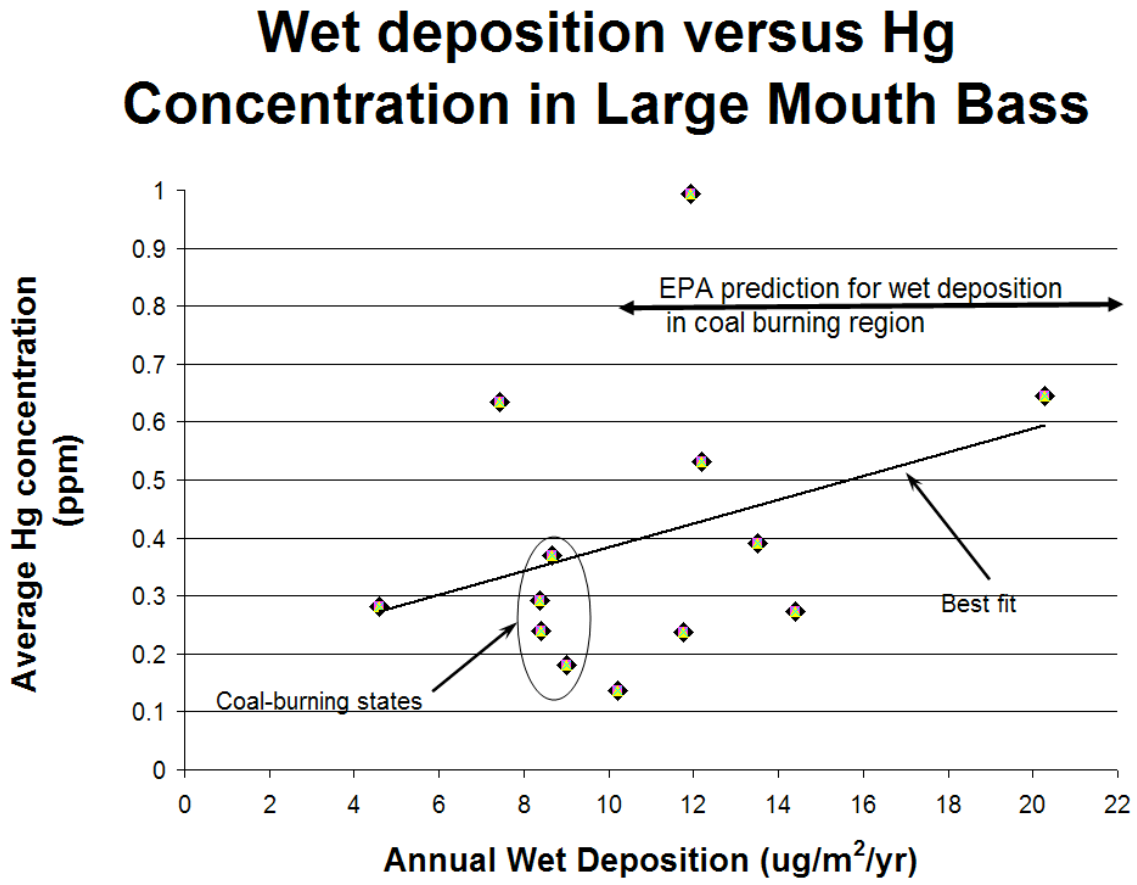

Figure C-2 Relationship between mercury in large moth bass and wet deposition of Hg,based on ssstate-level data.

Downwind Mercury Measurements Show Reductions of Reactive Gaseous Mercury (RGM) to Elemental Hg (Hg_0) in Coal-Fired Power-Plant Plumes

Eric Edgerton, Ben Hartsell, and John Jansen

Llubljana, Slovenia, July 2, 2004. SO_2 was used as a tracer in experimental studies of mercury chemistry and dilution in several southeastern coal-fired power-plant plumes. Emissions of SO_2, RGM, and Hg_0 were used to predict the values that would be expected to be seen in downwind ground-level measurements. These experiments were made in the absence of precipitation, and conservation of emitted mass suggests accurate measurements. These data show an average RGM loss of about 70% with a similar gain in Hg_0, over several years of observations.

Presented at the 7th International Conference on Mercury as a Global Pollutant

Our Comment: Atmospheric modeling results (shown in Figure C-3 indicate that a 2/3 reduction of RGM to Hg(0) as it leave a coal-fired power plant removes most of the "Pennsylvania anomaly", providing additional ground-truth validation Thus, changes to the atmospheric chemistry modules used in national and global modeling programs appear to be in order.

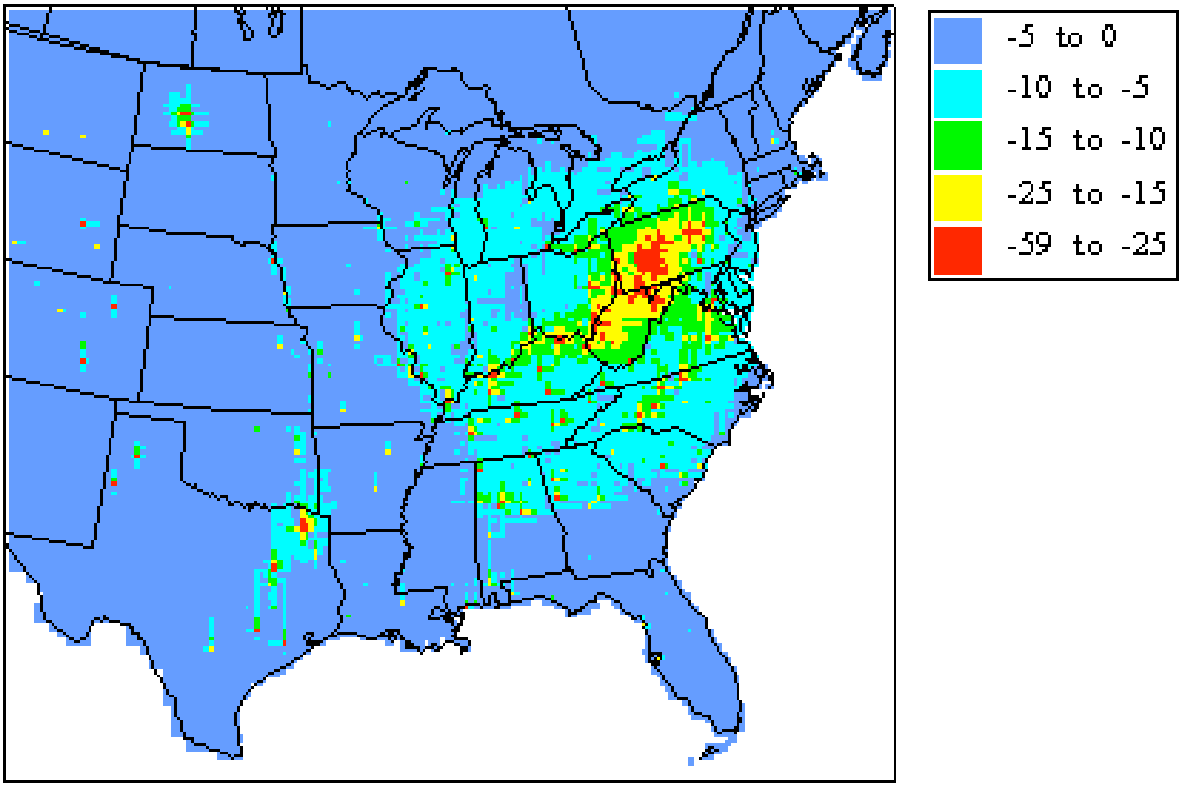

Figure C-3. Changes in Total Hg Deposition Due to Reduction of Reactive Gaseous Mercury Emissions to $Hg_{(0)}$ Emissions from Coal-fired Power Plants by 67%

Source: "Modeling the Impact of Mercury Speciation in Power Plant Plumes on Hg Deposition over the Eastern United States, K. Vijayaraghavan, C. Seigneur, K. Lohman, P. Karamchandani, L. Levin, J. Jansen. Presented at the 7th Annual Electric Utilities Environmental Conference, Tucson, AZ January 20-22, 2004.

Recent Publications Find Excess Hg Deposition in Urban Areas

While many of the concerns about mercury in the environment focus on remote and relatively pristine areas like the Arctic, several relatively recent publications in the atmospheric literature focus on urban areas:

- Hg particles washed from rooftops (Chemosphere, Volume 52, pp. 1727-41, 2003).
- Chicago-Gary urban area (Environmental Sci & Tech., Vol. 36, pp. 4508-17, 2002).
- Connecticut (Chemosphere, Volume 45, pp. 1033-43, 2001).
- Syracuse, NY (Neurotoxicology, Volume 17, pp. 279-90, 1996).

Our Comment: Because of the much higher urban population densities, contamination of fish taken from urban waters may warrant more attention than local deposition effects around remote power plants.

OUR BOTTOM LINE:

Models must be validated to be sufficiently credible to serve as a basis for public health policy. This will require detailed consideration of the reduction of RGM to Hg_0 in plumes from coal-fired power-plants and better data on rates of dry deposition. Additional data on ground truth are also needed, especially in the vicinities of power plants before and after mercury emission controls are installed. Realistic estimates of the atmospheric burdens of Hg from local sources are needed to allow effective control strategies to be devised.

C.5 Reviews, Editorials, and Opinions

Reviews of Health Effects

- ***Neurotoxic and Molecular Effects of Methyl mercury in Humans***
  Anna Castoldi, Teresa Coccini, and Luigi Manzo; University of Pavia, Italy.
  Reviews on Environmental Health, Volume 18, pp. 19-26 (2003).

"On the basis of epidemiological studies performed in populations having moderate chronic methyl mercury exposure, no definite consensus has been reached to date on the safety level of maternal exposure during pregnancy."

- ***Prenatal methyl mercury exposure and developmental effects. An evidence-based review.***
  Anne Spurgeon, University of Birmingham, United Kingdom.
  Presented at the 7th Int'l Conference on Mercury As a Global Pollutant, Slovenia, July 2004.

"The overall finding of the review therefore is that currently available evidence is unable to either support or refute the existence of an association between prenatal exposure to MeHg, derived from a maternal fish diet, and adverse developmental effects."

- ***Developmental neurotoxicity following prenatal exposures to methyl mercury and PCBs in humans from epidemiological studies***
  K Nakai and H Satoh, Tohoku University Graduate School of Medicine, Sendai, Japan.
  Tohoku Journal of Experimental Medicine, Volume 196, pp. 89-98 (2002).

"Since a considerable number of pollutants, including PCBs and pesticides, are also present in fish, … the combined effects of these pollutants should be considered in discussing the neurotoxicity of MeHg."

- ***Environmental factors associated with a spectrum of neurodevelopmental deficits***
  P Mendola, S Selevan, S Gutter, D Rice, U.S. EPA, Research Triangle Park, NC.
  Mental Retardation and Developmental Disability Research Review, Volume 8, pp. 188-97 (2002).

"Exposure to environmental agents with neurotoxic effects can result in a spectrum of adverse outcomes from severe mental retardation and disability to more subtle changes in function **depending on the timing and dose of the chemical agent**." (emphasis added).

- ***Evaluation of Uncontrolled Confounding in Studies of Environmental Exposures and Neurobehavioral Testing in Children***
  Pamela Mink, Michael Goodman, Leila Barraj, Harriet Imrey, Michael Kelsh, Janice Yager
  Epidemiology, Volume 15, pp. 385-393 (2004).

"Relatively small differences (0.5 standard deviations) in confounding variables between 'exposed' and 'unexposed' groups, if unmeasured and unaccounted for in the analysis, could produce spurious differences in cognitive test scores. The magnitude of this difference (3-10 points) has been suggested to have a meaningful impact in populations."

- ***Contending with contradictory data in a risk assessment context: the case of methyl mercury***
  Joseph Jacobson, Wayne State University, Detroit, MI.

Neurotoxicology, Volume 22, pp. 667-675 (2001).

"Because prospective epidemiological studies are often hampered by limited control over confounding and other factors, including unmeasured between cohort differences in genetic vulnerability and nutritional adequacy, inferences about toxicity often depend heavily on a qualitative assessment of the weight of evidence from multiple studies."

Our Comments: All of these reviews support the conclusion that no single study should be selected for public health policy and that a new American MeHg epidemiological study is needed.

Editorializing

- ***The Mercury Scandal***

Paul Krugman, New York Times, p. A-23, April 6, 2004.

"Sulfur dioxide is light, and travels long distances; power plants in the Midwest can cause acid rain in Maine. So a cap on total national emissions makes sense. Mercury is heavy; much of it precipitates to the ground near the source. As a result, coal-fired power plants in states like Pennsylvania and Michigan create "hot spots" – chemical Chernobyls – where the risks of mercury poisoning are severe. … That probably means thousands of children will be born with preventable neurological problems."

- ***Grappling with Mercury***

Jeff Johnson, Chemical and Engineering News, pp. 19-20, July 12, 2004.

"EPA estimates that some 630,000 children are borne each year with learning deficits caused by mercury exposure." "Critics also worry that the (EPA regulatory) proposals do not address mercury 'hot spots', where high mercury concentrations near emission sources have been found."

Our Comments: These two excerpts from publications of national and even international repute are egregious enough to raise questions about the entire print journalism industry. Paul Krugman seems to think that metallic mercury is emitted from smokestacks, and both he and Jeff Johnson apparently accept the existence of Hg "hot spots" near power plants without demanding proof on the ground. With respect to developmental impacts, a crucial qualifier has been ignored; EPA describes the impacts as children "at risk", which means their exposures are within the factor of 10 safety zone. It does not imply that the alleged neurological defects will actually occur. Further, the types of neurological effects observed above the factor of 10 safety zone are in the realm of a few (probably less than 1) IQ points, which hardly constitutes a "Chernobyl" situation.

Opinions (Correspondence and Commentary)

- ***Exceeding the Methyl Mercury Reference Dose: How Dangerous Is It?***

Deborah Schoen, Consultant/Science Writer, Longueuil, Quebec, Canada.

Environmental Health Perspectives, Volume 112, p. A337, 2004 (correspondence).

This letter refers to the research article by Jane Hightower (volume 111, pp. 604-608, 2003), which described Hg blood levels in a group of general practice patients, some of which had complained of various symptoms that might have been ascribed to mercury poisoning. The journal's cover carried the headline, "High Health Costs of Eating Expensive Fish," thus

implying that the paper was about health effects. In fact, Hightower specifically discounted any claims of finding cause-and-effect relationships; the article was only about exposures, and made frequent reference to EPA's guideline for pregnant females, even though only about _ of the subjects were pregnant. In her letter, Schoen stated "Because of this (Seychelles vs. Faeroes) uncertainty, the determination of the appropriate level for the methyl mercury RfD represents a subjective policy decision, as well as a calculation based on scientific data." In her response, Hightower listed some known health effects of mercury exposure but made no attempt to explain the uncertainty factor of 10 or the journal's mischaracterization of her paper.

- ***Do recent data from the Seychelles Islands alter the conclusions of the NRC report on the toxicological effects of methyl mercury?***

Alan Stern, Joseph Jacobson, Louise Ryan, Thomas Burke (members of the NRC Committee) Environmental Health, Volume 3, p.2, 2004 (commentary).

Stern et al. consider aspects of the Seychelles study and correspondence following a recent publication that found no adverse effects and conclude that none of these findings persuade them to consider that study in trying to establish a "safe" exposure level for the United States. "In the interest of protecting public health, we believe it is better to err on the side of caution in the face of three well-designed studies, two of which are positive (Faeroes, New Zealand) and one of which is negative (Seychelles)." … "Once reasonable evidence of adverse effects has been provided, the issue is not whether methyl mercury exposure from fish can pose a risk, but rather the dose (with an appropriate margin of safety) that is appropriate to provide prudent protection for the most vulnerable individuals in the population."

Our Comment: What Stern et al. do not consider is the effect of parallel exposures to PCBs in the Faeroes and the differences in exposure measures and statistical methods between the two studies. Furthermore, by selecting the most sensitive study and then imposing a safety factor of 10, they have effectively imposed two layers of caution.

Our Bottom Line: The lay literature is fraught with misconceptions and inaccuracies. There are large differences between assessments made by independent scientists and by those who have a regulatory agenda. The public interest is not well served by creating unwarranted fears about the safety of the food supply, no matter how worthy the environmental objective might be.